https://doi.org/10.5194/essd-2024-96
Preprint. Discussion started: 2 April 2024

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$PM_{2.5}$ concentrations based on near-surface visibility at 4011 sites in the Northern Hemisphere from 1959 to 2022

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Abstract

Long-term $PM_{2.5}$ data are needed to study the atmospheric environment, human health, and climate change. $PM_{2.5}$ measurements are sparsely distributed and of short duration. In this study, daily $PM_{2.5}$ concentrations are estimated from 1959 to 2022 using a machine learning method at 4011 terrestrial sites in the Northern Hemisphere based on hourly atmospheric visibility data, which are extracted from the Meteorological Terminal Aviation Routine Weather Report (METAR). $PM_{2.5}$ monitoring is the target of machine learning, and atmospheric visibility and other related variables are the inputs. The training results show that the slope between the estimated $PM_{2.5}$ concentration and the monitored $PM_{2.5}$ concentration is 0.946 ± 0.0002 within the 95% confidence interval (CI), the coefficient of determination (R^2) is 0.95, the root mean square error (RMSE) is 7.0 $\mu g/m^3$, and the mean absolute error (MAE) is 3.1 $\mu g/m^3$. The test results show that the slope between the predicted $PM_{2.5}$ concentration and the monitored $PM_{2.5}$ concentration is 0.862 ± 0.0010 within a 95% CI, the R^2 is 0.80, the RMSE is 13.5 $\mu g/m^3$, and the MAE is 6.9 $\mu g/m^3$. The multiyear mean $PM_{2.5}$ concentrations from 1959 to 2022 in the United States, Canada, Europe, China, and India are 11.2 $\mu g/m^3$, 8.2 $\mu g/m^3$, 20.1 $\mu g/m^3$, 51.3 $\mu g/m^3$ and 88.6 $\mu g/m^3$, respectively. $PM_{2.5}$ is low and continues to decrease from 1959 to 2022. $PM_{2.5}$ in the United States increases slightly at a rate of 0.38 $\mu g/m^3$/decade from 1959 to 1990 and decreases at a rate of -1.32 $\mu g/m^3$/decade from 1991 to 2022. Trends in Europe are positive (5.69 $\mu g/m^3$/decade) from 1959 to 1972 and negative (-1.91 $\mu g/m^3$/decade) from 1973 to 2022. Trends in China and India are increasing (3.04 and 3.35 $\mu g/m^3$/decade, respectively) from 1959 to 2012 and decreasing (-38.82 and -42.84 $\mu g/m^3$/decade, respectively) from 2013 to 2022. The dataset is available at National Tibetan Plateau / Third Pole Environment Data Center (https://doi.org/10.11888/Atmos.tpdc.301127) (Hao et al., 2024).

Keywords

Fine particulate matter; $PM_{2.5}$; Visibility; Machine learning; Dataset.

1 Introduction

Fine particulate matter ($PM_{2.5}$) refers to particulate matter suspended in air with an aerodynamic diameter of less than 2.5 micrometers. $PM_{2.5}$ has various shapes and is composed of complex components, such as inorganic salts (e.g., sulfate, nitrate, and ammonium), as well as organic carbon

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and elemental carbon, metallic elements, and organic compounds (Chen et al., 2020; Fan et al., 2021). $PM_{2.5}$ can be emitted directly into the atmosphere (Viana et al., 2008; Zhang et al., 2019) and generated through photochemical reactions and transformations (Guo et al., 2014). $PM_{2.5}$ exhibits high concentrations near emission sources, which gradually decreases with distance. Due to the small size and longer life span of $PM_{2.5}$ compared with coarse particulate matter, it can be transported over long distances by atmospheric movements, leading to wide-ranging impacts. Studies indicate that regional transport contributes significantly to local $PM_{2.5}$ (Wang et al., 2014; Chen et al., 2020).

$PM_{2.5}$ reduces atmospheric visibility and facilitates the formation of fog and haze conditions (Fan et al., 2021). Direct and indirect effects on solar radiation in the atmosphere (Albrecht, 1989; Ramanathan et al., 2001; Bergstrom et al., 2007; Chen et al., 2022) alter the energy balance and the number of condensation nuclei, thereby influencing atmospheric circulation and the water cycle (Wang et al., 2012; Liao et al., 2015; Samset et al., 2019; Li et al., 2022).

$PM_{2.5}$ is also known as respirable particulate matter. Due to its complex composition, $PM_{2.5}$ may carry toxic substances that can significantly impair human health. The World Health Organization states explicitly that $PM_{2.5}$ is more harmful than coarse particles, and long-term exposure to high $PM_{2.5}$ concentrations increases the risk of respiratory diseases, cardiovascular diseases, and lung cancer (Lelieveld et al., 2015), regardless of a country's development status. A Global Burden of Diseases study revealed that exposure to environmental $PM_{2.5}$ causes thousands of deaths and millions of lung diseases annually (Chafe et al., 2014; Kim et al., 2015; Cohen et al., 2017).

$PM_{2.5}$ is an important parameter for assessing particulate matter pollution and air quality (Wang et al., 2012). $PM_{2.5}$ can lead to soil acidification, water pollution, disruption of plant respiration, and ecological degradation (Wu and Zhang, 2018; Liu et al., 2019). Due to globalization and economic integration, preventing and controlling particulate matter pollution is a challenge at city, country and global scales.

Therefore, long-term $PM_{2.5}$ data are needed for studies on the environment, human health, and climate change. At present, ground-based measurements, chemical models, and estimations of alternatives are the primary sources of $PM_{2.5}$ data.

Ground-based measurements are the most effective means to measure $PM_{2.5}$. $PM_{2.5}$ monitoring has been ongoing since the 1990s in North America and Europe (Van Donkelaar et al., 2010), and large-scale $PM_{2.5}$ monitoring has been implemented in other regions since 2000, including China in 2013 (Liu et al., 2017). As a result, the records for $PM_{2.5}$ are short, with only a few years of data available in many countries. The scarcity of $PM_{2.5}$ measurements makes it challenging to provide long-term historical data for research.

Reanalysis datasets provide estimates of long-term particulate matter concentrations. The Modern-Era Retrospective Analysis for Research and Applications version 2 (MERRA-2) is a reanalysis dataset from NASA that uses the Goddard Earth Observing System version 5 (GEOS-5), which has provided global $PM_{2.5}$ data since 1980 (Buchard et al., 2015; Buchard et al., 2016; Buchard et al., 2017; Gelaro et al., 2017; Sun et al., 2019). The MERRA-2 surface $PM_{2.5}$ assessment results are more consistent between observations located in rural areas, as cities and suburban areas are affected by high local emissions that do not represent the estimated grid average. Due to the lack of nitrate

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and low organic carbon emissions in GOCART, there is a difference in the total amount of $PM_{2.5}$ during winter in the western United States, and sea salt aerosols are overestimated (Buchard et al., 2017). Another reanalysis dataset is the Copernicus Atmosphere Monitoring Service (CAMS) global reanalysis, which is a global reanalysis dataset of the atmospheric composition produced by the European Centre for Medium-Range Weather Forecasts (ECMWF) and has provided $PM_{2.5}$ data since 2003 (Che et al., 2014; Inness et al., 2019). The validation of $PM_{2.5}$ for CAMS shows severe overestimations in some areas (Ali et al., 2022; Jin et al., 2022). Although reanalysis provides long-term $PM_{2.5}$ data, the uncertainty in emission inventories increases the uncertainty in $PM_{2.5}$, which remains challenging (Granier et al., 2011).

Many studies have employed statistical methods, machine learning, and deep learning methods to estimate $PM_{2.5}$ concentrations based on aerosol optical depth (AOD). Van Donkelaar et al. (2021) utilized satellite AOD, chemical transport models, and ground-level measurements of AOD to estimate monthly $PM_{2.5}$ concentrations and their uncertainties over global land from 1998 to 2019, and there are several related studies (Van Donkelaar et al., 2010; Boys et al., 2014; Van Donkelaar et al., 2015; Van Donkelaar et al., 2016; Hammer et al., 2020). Many studies have been conducted at the regional scale, such as in the United States (Beckerman et al., 2013), China (Wei et al., 2019b; Xue et al., 2019; Wei et al., 2020a; He et al., 2021; Wei et al., 2021), and India (Mandal et al., 2020). Although the $PM_{2.5}$ data derived from satellite retrievals have high spatial coverage, the temporal range depends entirely on the satellite retrievals. The estimation of $PM_{2.5}$ based on satellite products is also limited by bright surfaces, cloud conditions (Wei et al., 2019a) and resolution (Nagaraja Rao et al., 1989; Hsu et al., 2017).

Another alternative for estimating $PM_{2.5}$ concentrations is the atmospheric horizontal visibility, which is the maximum distance at which observers with normal visual acuity can discern target contours under current weather conditions. In addition to manual observations, automated visibility measurements were implemented early, typically relying on the aerosol scattering principle (Wang et al., 2009; Zhang et al., 2020). Visibility and $PM_{2.5}$ are measurements of near-surface aerosols. They describe atmospheric transparency and are used to describe atmospheric pollution. Long-term visibility records have been used to quantify long-term aerosol properties (Molnár et al., 2008; Wang et al., 2009; Zhang et al., 2017; Zhang et al., 2020). Visibility observation stations are densely distributed across the country. Compared to satellite-retrieved AOD data, visibility observations have longer historical records dating back to the early 20th century (Noaa et al., 1998; Boers et al., 2015), are not affected by cloud interference and provide continuous measurements.

Visibility has been used as a proxy for $PM_{2.5}$ (Huang et al., 2009) and to estimate $PM_{2.5}$ (Liu et al., 2017; Li et al., 2020; Singh et al., 2020). Singh et al. (2020) analyzed the air quality in East Africa from 1974 to 2018 using visibility data. Liu et al. (2017) developed a statistical model and utilized ground-level visibility data to estimate long-term $PM_{2.5}$ concentrations in China from 1957 to 1964 and 1973 to 2014. Gui et al. (2020) proposed a method to establish a virtual ground observation network for $PM_{2.5}$ in China using extreme gradient boosting modeling in 2018. Zeng et al. (2021) used LightGBM to establish a virtual network for hourly $PM_{2.5}$ concentrations in China in 2017. Zhong et al. (2021; 2022) used LightGBM to predict 6-hour $PM_{2.5}$ concentrations based on visibility, temperature, and relative humidity in China from 1960 to 2020. Meng et al. (2018) utilized a random forest model to estimate the daily $PM_{2.5}$ components in the United States from 2005 to 2015. These studies have provided various methods for estimating $PM_{2.5}$ using visibility data. However, some

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have focused on only methodological innovations without providing long-term trends in $PM_{2.5}$. Other studies offer long-term trends, but the primary focus was at urban and national scales. There are few studies on long-term and high-temporal-resolution $PM_{2.5}$ at the global scale or across different countries.

This study uses a convenient, accurate, and easily understandable machine learning approach to estimate daily $PM_{2.5}$ concentrations based on visibility at 4,011 land-based sites from 1959 to 2022. We also provide the long-term trends and characteristics of $PM_{2.5}$ in different regions. The $PM_{2.5}$ dataset provides support for climate change, human health, and pollution control research. First, we build a machine learning model and then analyze the importance of the variables. Second, we evaluate the model's performance and predictive ability. Third, we discuss the errors and limitations of the dataset. Fourth, we compare the estimated $PM_{2.5}$ with the other datasets. Finally, we analyze the spatial-temporal distributions of $PM_{2.5}$.

2 Data and methods

2.1 Study Area

The study area includes Canada, the United States, Europe, China, and India in the Northern Hemisphere. The distributions of visibility stations (a) and the $PM_{2.5}$ monitoring sites (b) in each region are shown in Figure 1. The number of visibility stations is 3177, and a total of 4011 $PM_{2.5}$ monitoring sites are selected for this study, with 1110 sites in the United States, 304 sites in Canada, 834 sites in Europe, 1557 sites in China, and 206 sites in India.

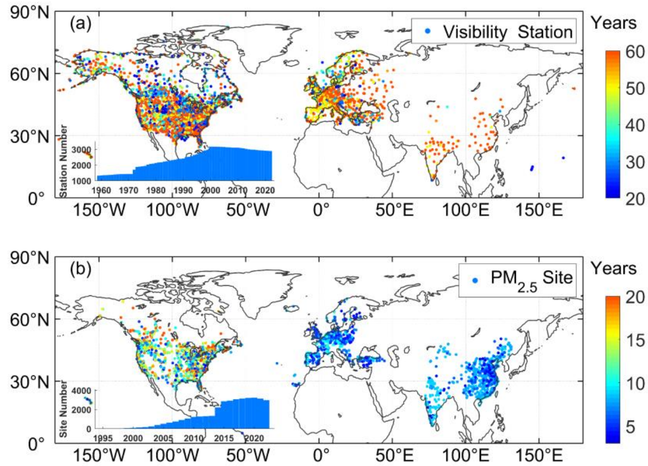

Figure 1 Study area and the distribution of visibility stations from 1959 to 2022 (a) and $PM_{2.5}$ monitoring sites from 1995 to 2022 (b). The color of marker (circle) represents that the length of the observation record of visibility and $PM_{2.5}$ observations. The bar chart shows the number of visibility stations and $PM_{2.5}$ monitoring sites per year. The number of visibility stations is 3177.The number of $PM_{2.5}$ sites is 4011 in this study (1110 in the United States, 304 in Canada, 834 in Europe, 1557 in China, and 206 in India).

2.2 $PM_{2.5}$ Data

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2.2.1 $PM_{2.5}$ Data in the United States

The hourly $PM_{2.5}$ data for the United States from 1998 to 2022 are sourced from the Air Data System (AQS), which are available at https://www.epa.gov/aqs. The AQS provides $PM_{2.5}$ mass monitoring and routine chemical speciation data and contains other ambient air pollution data collected by the Environmental Protection Agency (EPA), state, local, and tribal air pollution control agencies from thousands of monitors, comprising the Federal Reference Method (FRM) and Federal Equivalent Method (FEM). The primary purpose of both methods is to assess compliance with the $PM_{2.5}$ National Ambient Air Quality Standards (NAAQS). FRMs include in-stack particulate filtration, and FEMs include beta-attenuation monitoring, very sharp cut cyclones, and tapered element oscillating microbalances (TOEMs). The measurement precision is ± (1~2) $\mu g/m^3$ (hour) (Gilliam and Hall, 2016). The TEOM and beta-attenuation are automatic and near real-time monitoring methods. The TEOM, which is based on gravity, measures the mass of particles collected on filters by monitoring the frequency changes in tapered elements. The beta-attenuation method uses beta-ray attenuation and particle mass to measure the $PM_{2.5}$ concentration. In this study, we use two $PM_{2.5}$ measurement methods, FRM/FEM (88101) and non-FRM/FEM (88502). The 88502 monitors are "FRM-like" but are not used for regulatory purposes. Both the 88101 and 88502 monitors are used for reporting daily Air Quality Index values.

We set the conditions that each $PM_{2.5}$ monitoring event have a minimum of 3 years and more than 1000 days of overlapping records with nearby visibility stations. A total of 1110 sites in the United States are selected for this study.

2.2.2 $PM_{2.5}$ Data in Canada

The hourly $PM_{2.5}$ data for Canada from 1995 to 2022 are sourced from the National Air Pollution Surveillance (NAPS) program, which are available at https://www.canada.ca. The NAPS program is a collaborative effort between the Environment and Climate Change Canada and provincial, territorial, and regional governments and is the primary source of environmental air quality data. Since 1984, $PM_{2.5}$ concentrations have been measured in Canada using a dichotomous sampler. Continuous or real-time particle monitoring began in the NAPS network in 1995 using TEOM and beta-attenuation monitoring (Demerjian, 2000). The samples are supplemented by EPA FRM samples obtained after 2009 (Dabek-Zlotorzynska et al., 2011). The number of instruments is growing rapidly, with 410 sites in 2022. A total of 304 $PM_{2.5}$ monitoring sites in Canada are selected for this study.

2.2.3 $PM_{2.5}$ Data in Europe

The hourly $PM_{2.5}$ data for Europe from 1998 to 2012 are obtained from the AirBase database, which is available at https://european-union.europa.eu. The hourly $PM_{2.5}$ verified data (E1a) from 2013 to 2022 are obtained from the AirQuality database, which is available at https://www.eea.europa.eu. AirBase is maintained by the European Environment Agency (EEA) through its European Topic Center on Air Pollution and Climate Change Mitigation. Airbase contains air quality monitoring data and information submitted by participating countries throughout Europe. After the Air Quality Directive 2008/50/EC was enforced, the $PM_{2.5}$ data began to be stored in AirQuality database. The main monitoring methods for $PM_{2.5}$ include TEOM and beta attenuation (Green and Fuller, 2006; Chow et al., 2008). The sites are distributed across rural, rural-near city, rural-regional, rural-remote,

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suburban, and urban areas. We merge the two datasets with the same site identifiers, and 834 sites in Europe are selected for this study.

2.2.4 $PM_{2.5}$ Data in China

The hourly $PM_{2.5}$ data for China from 2014 to 2022 are obtained from the China National Environmental Monitoring Center, which are available at https://www.cnemc.cn. China established air quality monitoring in 1980; 74 cities were the first to publicly release real-time $PM_{2.5}$ in 2013, and there were more than 1800 air quality observation sites as of 2000 (Su et al., 2022). $PM_{2.5}$ concentrations are measured using the TEOM and beta-attenuation method (Zhao et al., 2016b; Miao and Liu, 2019). According to the China Environmental Protection Standards, instrument maintenance, data transmission, data assurance and quality control ensure the reliability of $PM_{2.5}$ concentration measurements. The uncertainty in the $PM_{2.5}$ mass concentration is <5 μg/m^{-3} (Pui et al., 2014). In this study, a total of 1110 $PM_{2.5}$ monitoring sites are selected.

2.2.5 $PM_{2.5}$ Data in India

The hourly $PM_{2.5}$ data for India from 2010 to 2022 are obtained from the Central Pollution Control Board (CPCB), which are available at https://app.cpcbccr.com. The Air (Prevention and Control of Pollution) Act of 1981 was enacted by the Central Pollution Control Board (CPCB) of the Ministry of Environment, Forest and Climate Change (MoEFCC). A standard of 60 μg/m^3 $PM_{2.5}$ concentration over 24 hours was added in 2009. The methods used by the Indian National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$ and related component measurements include the TEOM, FRM and FEM (Pant et al., 2019). The measurement precision is ± (1-2) μg/m^3 (hour). The National Air Quality Monitoring Programme (NAMP) is a key air quality monitoring programme employed by the Government of India, which is managed by the CPCB in coordination with the State Pollution Control Boards (SPCBs) and UT (union territory) Pollution Control Committees (PCCs). There were 703 $PM_{2.5}$ monitoring stations as of 2018. Most of these stations (residential and industrial) are located in urban areas, and others are located sparsely in rural areas. A total of 206 $PM_{2.5}$ monitoring sites are selected for this study.

2.3 Visibility and Meteorological Data

The hourly meteorological data from 1959 to 2022 are collected from airport weather observations, which are available at https://www.weather.gov/asos. Automated observation minimizes the errors associated with human involvement in data collection, processing, and transmission. The data are extracted from the Meteorological Terminal Aviation Routine Weather Report (METAR). The World Meteorological Organization (WMO) sets guidelines for METAR reports, including report format, encoding, observation instruments and methods, data accuracy, and consistency. These requirements ensure the consistency and comparability of METAR reports globally. Visibility is a quantity that describes the atmospheric transparency, usually observed by automated sensors (scattering and transmission). More than 1000 stations are from the Automated Surface Observing System (ASOS) in the United States, and other data are sourced from airport reports worldwide. The forward-scatter visibility sensors at a wavelength of 550 nm for ASOS are consistent with the National Weather Service of the United States standard transmissometer, with more than 80% of the data within the limit of ±0.4 km when visibility is less than 2 km (Noaa et al., 1998).

Visibility is an essential variable employed in this study, as research has shown that its reciprocal is

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directly proportional to the aerosol extinction coefficient (Wang et al., 2009), which is closely related to the $PM_{2.5}$ concentration. Considering that temperature, wind speed, wind direction, humidity, and precipitation are factors that impact particle dispersion, particle growth, and secondary generation influenced by humidity, as well as the cleansing effect of precipitation (Zhang et al., 2020), temperature, dew point temperature, temperature-dew point difference, relative humidity, sea-level pressure, wind speed and direction, precipitation, and sky conditions are also employed in this study.

2.4 Data Preprocessing

The following data preprocessing steps are performed: remove the records with missing visibility, temperature, dew point temperature, temperature-dew point difference, relative humidity, sea-level pressure, wind speed, and wind direction data and remove records with hourly precipitation greater than 0.1 mm, sky conditions marked as 'VV', and relative humidity greater than 90%. Since $PM_{2.5}$ exhibits hygroscopic growth, we calculated the dry visibility for relative humidity values between 30% and 90% (Yang et al., 2021).

$$\boldsymbol{VISD = VIS/(0.26 + 0.4285 * log(100 - RH))}$$

where VIS is the visibility, RH is the relative humidity, and VISD is the dry visibility.

The maximum hourly $PM_{2.5}$ concentration is set to 1000 μg/m^3. At least three hourly daily records are needed. The harmonic mean is used to calculate the daily VIS and daily VISD because it can better capture rapid weather changes and enhance daily representativeness (Noaa et al., 1998). The arithmetic average is used for other variables.

2.5 Data for Comparison

In this study, our data are compared with other datasets, including two $PM_{2.5}$ datasets based on satellite AOD data and two reanalysis datasets.

2.5.1 ACAG Dataset

The monthly global $PM_{2.5}$ dataset (version V5.GL.04) from 1980 to 2022, with a spatial resolution of 0.1°, is available from the Atmospheric Composition Analysis Group (ACAG) of Washington University in St. Louis (https://sites.wustl.edu/acag/datasets/surface-pm2-5/) (Van Donkelaar et al., 2021). The ACAG $PM_{2.5}$ concentrations are estimated based on satellite (MODIS, VIIRS, MISR and SeaWiFS) AOD and global vertical aerosol profiles from the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) satellites. The AOD of GEOS-Chem is used to simulate the spatiotemporally varying geophysical relationship with $PM_{2.5}$. Ground-based $PM_{2.5}$ values are incorporated at a monthly timescale using geographically weighted regression (Van Donkelaar et al., 2016; Hammer et al., 2020; Van Donkelaar et al., 2021). The coefficients of determination (R^2) for the monthly mean and monitor-based $PM_{2.5}$ concentrations are 0.86 (January), 0.81 (April), 0.72 (July), and 0.78 (October). The R^2 with WHO-collocated monitors is between 0.88 and 0.93. The EMSE is between 8 and 13.3 μg/m^3.

2.5.2 CHAP Dataset

The monthly $PM_{2.5}$ dataset of China High Air Pollutants (CHAP) from 2000 to 2021 is a product with coverage over China, with a spatial resolution of 1 km, which is available at

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https://zenodo.org/records/6398971. The CHAP $PM_{2.5}$ concentration is estimated based on the MODIS Collection 6 MAIAC AOD product and meteorological variables, surface conditions, pollutant emissions, and population distributions using a space-time extra-trees model. The R^2 and RMSE of the monthly $PM_{2.5}$ concentration are 0.92-0.94 and ~5.1-10.0 μg/m^3, respectively, from 2013 to 2018 (Wei et al., 2020b; Wei et al., 2021).

2.5.3 MERRA-2 Dataset

The monthly $PM_{2.5}$ dataset of Modern-Era Retrospective Analysis for Research and Applications version 2 (MERRA-2) from 1980 to 2022 is a NASA reanalysis dataset with a spatial resolution of 0.5×0.625° and uses the Goddard Earth Observing System version 5 (GEOS-5) coupled to the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model, which is available at https://gmao.gsfc.nasa.gov. The aerosol data of GOCART include dust, sea salt, sulfate, black carbon, and organic carbon, and there are 72 vertical layers from the surface to more than 80 km altitude. MERRA-2 $PM_{2.5}$ is a dataset produced by the GEOS-5 atmospheric model and data assimilation system and the three-dimensional variational data analysis (3DVAR) Grid-point Statistical Interpolation (GSI) meteorological analysis scheme (Randles et al., 2017). In the aerosol model (GOCART), a SO_2 emission database of volcanic material for secondary sources is included. Aerosol hygroscopic growth depends on the simulated relative humidity. The monthly scale biomass burning inventory is from RETROv2 from 1980 to 1996; the monthly SO_2, SO_4, POM, and BC emissions are from GFEDv3.1 from 1997 to 2009; and the daily scale data are from QFED 2.4-r6 after 2010. The annual anthropogenic SO_2 is from EDGARv4.2 between 100 and 500 m above the surface from 1980 to 2008. The annual Anthropogenic SO_4, BC, and POM concentrations are obtained from AeroCom Phase II from 1980 to 2006. In assimilation systems, satellite AOD retrievals are used, including AVHRR (over the oceans) from 1998 to 2002, MISR from 2000 to 2014, MODIS Aqua since 2002, and MODIS Terra since 2000 (Buchard et al., 2017; Randles et al., 2017). The direct observations of the AOD AERONET station from 1999 to 2014 are also assimilated.

The surface $PM_{2.5}$ concentration in MERRA-2 can be computed using the concentrations of black carbon [BC], organic carbon [OC], dust [$DUST_{2.5}$], sea salt [$SS_{2.5}$], and sulfate [SO_4] (Provençal et al., 2017) and is expressed as follows (please refer to https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/FAQ/#Q4):

$$[PM_{2.5}] = [DUST_{2.5}] + [SS_{2.5}] + [BC] + 1.6\times[OC] + 1.375\times[SO_4].$$

In this study, we conduct spatiotemporal matching between MERRA-2 $PM_{2.5}$ and the estimated $PM_{2.5}$.

2.5.4 CAMS Dataset

The Copernicus Atmosphere Monitoring Service (CAMS) reanalysis is the latest global reanalysis dataset of atmospheric composition produced by the European Centre for Medium-Range Weather Forecasts (ECMWF). We use the single-level monthly $PM_{2.5}$ product from the CAMS reanalysis from 2003 to 2022, which is available at https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-reanalysis-eac4. The resolution is 0.75°. The CAMS reanalysis builds on the experience gained during the earlier Monitoring Atmospheric Composition and Climate (MACC) reanalysis and CAMS interim reanalysis (Inness

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et al., 2019). The ECMWF's Integrated Forecast System (IFS) aerosol and chemistry modules are applied, and more details on the modules are provided in (2015). The data at 60 model levels are interpolated to 25 pressure levels. Anthropogenic emissions are from the MACCity inventory from 1960 to 2010 (Granier et al., 2011). The emissions of anthropogenic SOAs are estimated from MACCity CO emissions. The monthly biogenic emissions of the chemical species are from MEGAN2.1 (Guenther et al., 2006). The natural NO_2 emissions from soils and oceans are obtained from the Precursors of Ozone and Their Effects in the Troposphere (POET) database for 2000. Daily biomass burning emissions are from the Global Fire Assimilation System version 1.2 (GFASv1.2) (Kaiser et al., 2012). More details regarding emissions are provided in Granier (2011). The incremental 4D-Var data assimilation system is used for the CAMS reanalysis, and the total aerosol mixing ratio of the single species is derived from the assimilation of satellite retrievals (Benedetti et al., 2009). The AODs from satellite retrievals are assimilated, including those from AATSR Envisat from 2002 to 2012 and those from MODIS Terra and Aqua since 2002. For additional information, please refer to Inness et al. (2019).

The surface $PM_{2.5}$ concentration is estimated by the air density [ρ], sea salt [$SS_{1,2}$], dust [$DD_{1,2,3}$], nitrate [$NI_{1,2}$], organic matter [OM], black carbon [BC], ammonium [AM], and sulfate [SO_4] and is expressed as follows (Inness et al., 2019):

$$[PM_{2.5}] = \rho \times([DD_1] + [DD_2] + [SS_1/4.3] + [0.5 \times SS_2/4.3] + [0.7 \times (AM + OM + 0.7NI_1 + SO_4)] + [BC] + 0.25 \times[NI_2]).$$

2.6 Decision Tree Regression

We employ decision tree regression using the CART algorithm (Teixeira, 2004) to estimate daily $PM_{2.5}$ concentrations. The key to decision tree regression is to find the optimal split variable and optimal split point. The optimal split point of the predictor is determined by the minimum mean squared error, which determines the optimal tree structure. Decision tree regression is a commonly used nonlinear machine learning method that partitions the feature space based on the mapping between feature attributes and response values, with each leaf node representing a specific output for each feature space region. It's ability to handle complex relationships with relatively few model parameters is advantageous, minimizing the risk of overfitting and enabling the prediction of continuous and categorical predictive variables.

The predictor includes 11 variables: the reciprocal of dry visibility (Vis_Dry_In), the reciprocal of visibility (Vis_In), temperature (Temp), dew point temperature (Td), temperature-dew point difference (Temp-Td), relative humidity (RH), sea-level pressure (SLP), wind speed (WS), wind direction (WD), numerical time (DateTime) and daily record number (DailyObsNum). The response variable is the daily observed $PM_{2.5}$ concentration.

We randomly select 80% of the sample data to establish the decision tree regression model, and the remaining 20% of the sample data are used to test the model's predictive ability. To obtain a stable model, a 10-fold cross-validation method (Browne, 2000) is used to train the model.

2.7 Evaluation Metrics

2.7.1 Statistical Metrics

We use the root mean squared error (RMSE), mean absolute error (MAE), and correlation

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coefficient (ρ) as evaluation metrics to evaluate the model's performance and predictive ability. The formulas are given as follows:

$$\boldsymbol{MSE} = \sqrt{\frac{\boldsymbol{1}}{\boldsymbol{n}}\sum_{\boldsymbol{i=1}}^{\boldsymbol{n}}(\boldsymbol{y_i} - \boldsymbol{\hat{y}_i})^{\boldsymbol{2}}}$$

$$\boldsymbol{MAE} = \frac{\boldsymbol{1}}{\boldsymbol{n}}\sum_{\boldsymbol{i=1}}^{\boldsymbol{n}}|\boldsymbol{y_i} - \boldsymbol{\hat{y}_i}|$$

$$\boldsymbol{\rho} = \frac{\sum_{i=1}^{n}(\boldsymbol{y_i} - \boldsymbol{\bar{y}})(\boldsymbol{\hat{y}_i} - \boldsymbol{\bar{\hat{y}}})}{\boldsymbol{sqrt}(\sum_{i=1}^{n}(\boldsymbol{y_i} - \boldsymbol{\bar{y}})^2 \sum_{i=1}^{n}(\boldsymbol{\hat{y}_i} - \boldsymbol{\bar{\hat{y}}})^2)}$$

where y_i and $\bar{y}$ are the predicted value and the average of the predicted values. $\hat{y}_i$ and $\bar{\hat{y}}$ are the target and the average of the target. $i = 1,2,\ldots,n$. n is the length of sample.

2.7.2 Partial Dependence

The importance of predictor variables is assessed via partial dependence. Partial dependence represents the relationship between the individual predictive variable and the predicted response (Friedman, 2001). By marginalizing the other variables, the expected response of the predicted variable is calculated. All the partial dependences of the predicted response on the subset of predicted variables are calculated. The calculation process of the partial dependency method is described as follows:

The dataset of the predictor is X, X $= [X^1, X^2, \ldots, X^n]$, and n represents the number of predictive factors. The complement of subset X^s is X^c, where X^s is a single variable in X and X^c is all other variables in X. The predicted response f(x) depends on all variables in X, and it is expressed as follows:

$$\boldsymbol{f}(\boldsymbol{x}) = \boldsymbol{f}(\boldsymbol{X^s}, \boldsymbol{X^c})$$

The partial dependence of the predicted response to X^s is expressed as follows:

$$\boldsymbol{f^s}(\boldsymbol{X^s}) = \int \boldsymbol{f}(\boldsymbol{X^s}, \boldsymbol{X^c})\boldsymbol{pC}(\boldsymbol{X^c})\boldsymbol{dX^c}$$

where $\mathrm{pC}(\mathrm{X^c})$ is the marginal probability of X^c, that is, $\mathrm{pC}(\mathrm{X^c}) \approx \int f(X^s, X^c)dX^s$. Assuming that the likelihood for each observation is equal, the dependence between X^s and X^c and the interactions of X^s and X^c in response are not strong. The partial dependence is shown below:

$$\boldsymbol{f^s}(\boldsymbol{X^s}) \approx \frac{\boldsymbol{1}}{\boldsymbol{N}}\sum_{\boldsymbol{i=1}}^{\boldsymbol{N}}\boldsymbol{f}(\boldsymbol{X^s}, \boldsymbol{X_i^s})$$

where N is the number of observations and $\boldsymbol{i}$ represents the ***i***th observation.

2.7.3 Mean Center

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The mean center is a geostatistical method used to describe the average position of a set of geographical coordinates. It represents the central tendency of a set of geographical data and aids in understanding the overall distribution and trends in the dataset. The mean center of the $PM_{2.5}$ concentration shows the overall trend and variability in $PM_{2.5}$. If the mean center is located at the edge of the dataset, the data distribution is dispersed. Conversely, if the mean center is located at the center of the dataset, the data distribution is concentrated. This may be relevant for aspects, such as population distribution, urban development, and economic activities. It is particularly helpful in understanding the spatial patterns of $PM_{2.5}$. The expression is given as follows:

$$\boldsymbol{x_{ct}} = \sum_{\boldsymbol{i=1}}^{\boldsymbol{N}} \boldsymbol{c_i} * \boldsymbol{x_i} / \sum_{\boldsymbol{i=1}}^{\boldsymbol{N}} \boldsymbol{c_i}$$

$$\boldsymbol{y_{ct}} = \sum_{\boldsymbol{i=1}}^{\boldsymbol{N}} \boldsymbol{c_i} * \boldsymbol{y_i} / \sum_{\boldsymbol{i=1}}^{\boldsymbol{N}} \boldsymbol{c_i}$$

where $\boldsymbol{x_{ct}}$ and $\boldsymbol{y_{ct}}$ represent the longitude and latitude of the mean center, respectively, and $\boldsymbol{c_i}$ represents the $PM_{2.5}$ concentration at the $\boldsymbol{i}$-th site ($\boldsymbol{x_i}$, $\boldsymbol{y_i}$).

2.7.4 Standard Deviation Ellipse

The standard deviation ellipse (SDE) is used in statistics and geography to describe the variability and correlation of multivariate data. The SDE is calculated based on the mean and covariance matrix of the data (Gong, 2002). This variable shows the dispersion and correlation of the data across different dimensions. The center of the ellipse corresponds to the mean of the data, while the shape and size of the ellipse reflect the variability in the data in different directions.

We calculate the SDE using the locations and concentration measurements associated with the $PM_{2.5}$ points. The major axis of the ellipse indicates the primary direction of data variation. The shape and size of the ellipse reflect the spatial dispersion of the $PM_{2.5}$ concentration. A larger ellipse indicates greater variability in the $PM_{2.5}$ concentration distribution, while a smaller ellipse denotes a more concentrated distribution. A circular ellipse indicates little or weak spatial correlation among $PM_{2.5}$ concentrations. A flattened ellipse indicates a spatial correlation between $PM_{2.5}$ concentrations.

3. Results and Discussion

3.1 Evaluation of Variable Importance

We analyze the influence of predictive variables over the predicted response. The predictive variable with the highest partial dependence value is the most important predictive variable in the model. The partial dependence of the predicted response on each predictive variable is calculated for every model. Figure 2 (a) shows the ranking results of the importance of all the predictive variables. The variable with the highest dependence on the predicted response is Vis_Dry_In, and the second highest dependence is Vis_In. The dependence of the predicted response on Temp, Td, Temp-Td, RH, WS, and wind WD is moderate. The predictive variables with lower dependence include SLP, DateTime and DailyObsNum.

We count the frequency and proportion of the most important variables in all the models, as shown

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in Figure 2 (b). Vis_Dry_In is the most important variable at 2600 sites, contributing 64.8%. Vis_In was the second most important variable at 575 sites, accounting for 14.3%. This finding indicates that visibility is the most crucial variable, with a percentage of 79.1%. Temp and Td contribute 6.7% and 3.5%, respectively. The contribution of other variables combined is 10.7%. The percentages of the second most important predictive variable are 25.4% for Vis_In, 39.6% for Vis_Dry_In, 14.6% for Temp, 7.1% for Td and 3.4% for Temp-Td. Among the three most important variables, the proportions of Temp and Td are 15.7% and 14.3%, respectively.

The results indicate a strong correlation between the $PM_{2.5}$ concentration and visibility, as visibility can be considered an indicator of air quality without fog or precipitation. Meteorological factors influence the dispersion and deposition of $PM_{2.5}$ (Gui et al., 2020; Zhong et al., 2022). Temperature and dew play secondary roles, and other meteorological predictive variables play lesser roles in the model. Although the number of daily records and time have the most negligible impacts on the $PM_{2.5}$ concentration in the model, they have significant impacts on the cyclical changes and daily representativeness of $PM_{2.5}$ (Wang et al., 2012; Zhang et al., 2020).

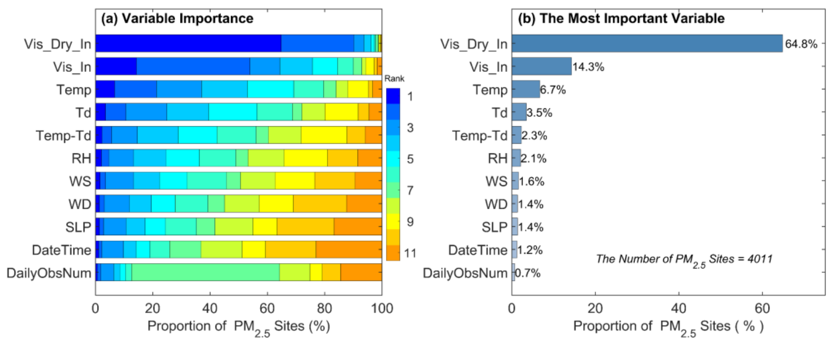

Figure 2 The importance of predictive variables. The stacked bar (a) shows the importance rankings of the predictive variables ('rank=1' represents the most important variable). The bar (b) shows the percentage of the most important predictive variable. The predictive variables are the reciprocal of dry visibility (Vis_Dry_In), reciprocal of visibility (Vis_In), temperature (Temp), dew point temperature (Td), temperature-dew point difference (Temp-Td), relative humidity (RH), sea level pressure (SLP), wind speed (WS), wind direction (WD), numerical time (DateTime) and daily record number (DailyObsNum). The total number of $PM_{2.5}$ sites is 4011.

3.2 Evaluation of Model Performance

3.2.1 For All Data

We analyze the linear fitting relationship between all estimated and corresponding response values to evaluate the model's performance. Figure 3 shows the density scatter plot of the monitored $PM_{2.5}$ concentration (response values) and the estimated $PM_{2.5}$ concentration (estimated values). There is a total of 8,680,796 data pairs for all the sites. The linear regression coefficient is 0.946 ±0.0002 within the 95% confidence interval, the R^2 is 0.95, the RMSE is 7.0 μg/m^3, and the MAE is 3.1 μg/m^3.

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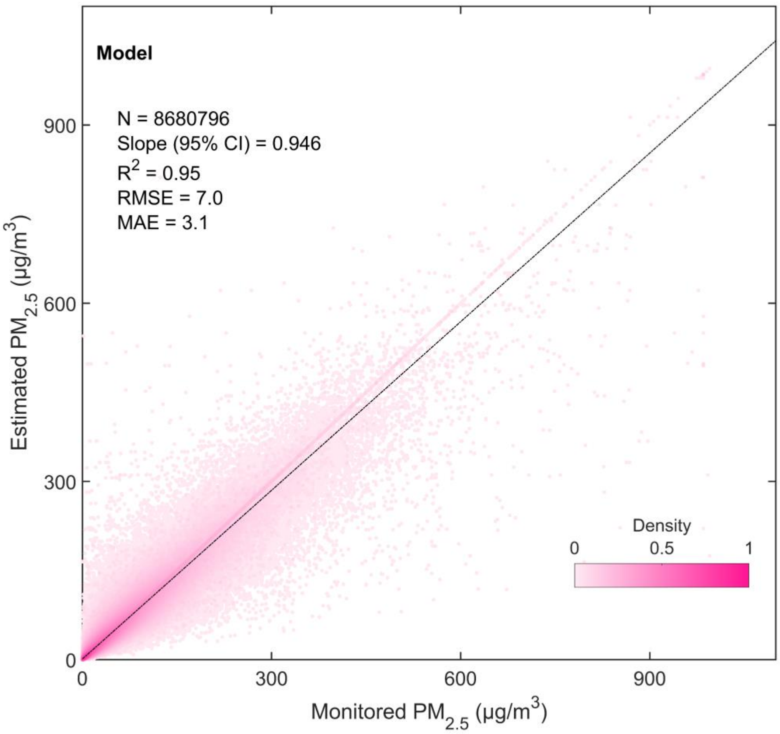

Figure 3 Density scatter plot (a) between estimated values (estimated $PM_{2.5}$) and the corresponding response values (monitored $PM_{2.5}$) at the daily scale. The dashed black line is the linear regression line. N is the length of the data pairs, and Slope is the linear regression coefficient within a 95% confidence interval (CI). R^2 is the coefficient of determination, RMSE is the root mean square error, and MAE is the mean absolute error.

3.2.2 For the Site and Region Scales

We evaluate the model's performance using the RMSE, MAE, and ρ of the estimated and response values at the site and region scales. Figure 4 shows the spatial distribution (a-c) and frequency distribution (d-f) of the model's RMSE, MAE, and ρ at all sites. Table 1 lists the model's performance metrics for all sites and sites in the United States, Canada, Europe, China, and India.

For all sites, the average RMSE is 7.42 μg/m^3, with a median of 4.97 μg/m^3. The RMSE of 80% of the sites is less than 11.95 μg/m^3. The ratio of the RMSE to the average $PM_{2.5}$ concentration is 29.2%. The average MAE is 4.01 μg/m^3, with a median of 2.66 μg/m^3. The MAE is less than 6.62 μg/m^3 for 80% of the sites. The MAE-to-mean ratio is 15.8%. The average ρ is 0.90, and the median is 0.91. The ρ of 80% of the sites is greater than 0.87. Previous studies have shown that for $PM_{2.5}$ retrieved from daily visibility or satellite AOD data, the R^2 range of the model is from 0.42 to 0.89, and the RMSE range is from 9.59 μg/m^3 to 32.09 μg/m^3 (Shen et al., 2016; Liu et al., 2017; Wei et al., 2019b; Gui et al., 2020; Li et al., 2021; Zhong et al., 2021). This finding indicates that our model

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performs well at the daily scale.

At the regional scale, the average RMSE values for the United States, Canada, Europe, China, and India are 78, 2.86, 4.63, 11.62, and 18.73 μg/m^3, respectively, and the mean $PM_{2.5}$ concentrations are 31.2%, 40.9%, 33.0%, 28.0%, and 27.9%, respectively. The average MAEs for the United States, Canada, Europe, China, and India are 1.42 μg/m^3, 1.36 μg/m^3, 2.45 μg/m^3, 6.48 μg/m^3, and 9.56 μg/m^3, respectively; these values correspond to 15.9%, 19.4%, 17.5%, 15.6%, and 14.2%, respectively, of the mean $PM_{2.5}$ concentration. The average correlation coefficients for the United States, Canada, Europe, China, and India are 0.88, 0.88, 0.89, 0.92, and 0.92, respectively.

The values of RMSE and MAE are the largest in India. The RMSE is the smallest in the United States, and the MAE is the smallest in Canada. The ratios of the RMSE and MAE to the mean are larger in Canada and Europe than in other regions and smaller in China and India than in other regions. Although the $PM_{2.5}$ concentration varies among regions, the MAE-to-mean concentration ratio remains at approximately 16%. This finding demonstrates the stability and reliability of the model.

Table 1 The results of the model's performance metrics for all sites and sites in the United States (the US), Canada, Europe, China and India.

Model	*RMSE (μg/m^3)*	*MAE (μg/m^3)*	*ρ (Pearson's correlation)*	*Mean (μg/m^3)*	*RMSE/Mean (%)*	*MAE/Mean (%)*
All	7.42	4.01	0.90	25.4	29.2	15.8
the US	2.78	1.42	0.88	8.9	31.2	15.9
Canada	2.86	1.36	0.88	7.0	40.9	19.4
Europe	4.63	2.45	0.89	14.0	33.0	17.5
China	11.62	6.48	0.92	41.5	28.0	15.6
India	18.73	9.56	0.92	67.0	27.9	14.2

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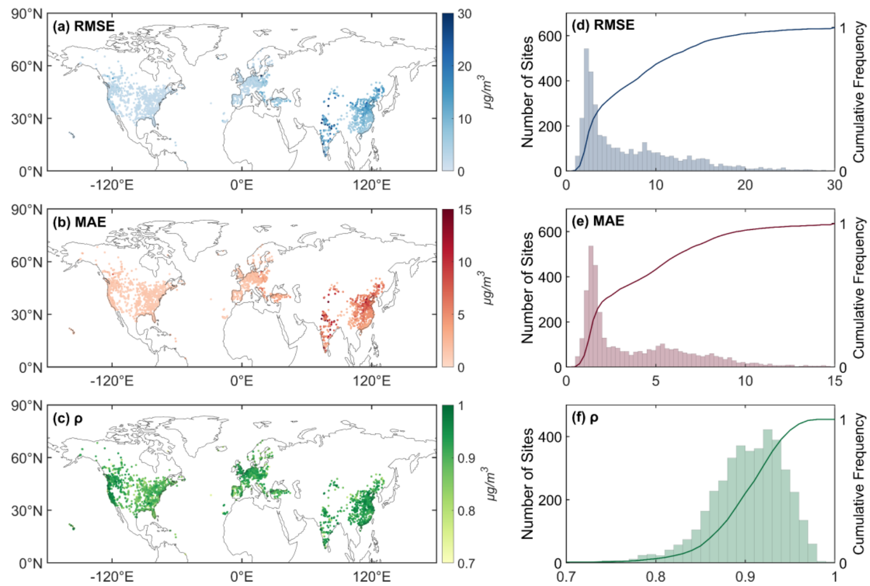

Figure 4 Spatial distribution (a-c) of the root mean squared error (RMSE), mean absolute error (MAE), and correlation coefficient (ρ) between the model's estimated values and response values. Number of sites (bar) and cumulative frequency (curve) (d-e) of the RMSE, MAE, and ρ.

3.2.3 Dependence on the Distance between the $PM_{2.5}$ Site and the Visibility Station

Although the previous analysis elucidates the stability and predictive capability of the model, it is necessary to understand the potential impact of the distance between $PM_{2.5}$ monitoring sites and visibility stations on the model. Most $PM_{2.5}$ monitoring sites are in urban areas, resulting in a relatively concentrated spatial distribution. Visibility stations are strategically placed to capture the characteristics of meteorological factors and have relatively uniform spatial distributions. Consequently, visibility stations and $PM_{2.5}$ monitoring sites are often not collocated, resulting in a certain spatial distance between them. Therefore, we consider the impact of the distance between sites on the model's performance.

Figure 5 shows the relationship between the model performance (ρ and RMSE) and the distance between the visibility stations and the $PM_{2.5}$ monitoring sites. The average distance between all sites is 0.964°, and the correlation coefficient between the model's RMSE and distance is 0.44, which is a moderate correlation. The average ρ of 3786 sites (within a distance of 3°) is 0.90, and the average RMSE is 7.13 μg/m³. The RMSE values of 471 sites are greater than twice the average RMSE of all sites; however, their average ρ (0.91) is greater than the average of all sites. This finding indicates that the model's performance decreases as the distance increases.

For the United States, the average distance is 0.29°. The distance between the 919 (82.8%) sites was less than 0.5°, with ρ and RMSE values of 0.88 and 2.7 μg/m³, respectively. The ρ and RMSE of the 191 sites (more than 0.5°) are 0.88 and 3.1 μg/m³, respectively. The performance of the model is not significantly related to distance.

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For Canada, 212 (69.7%) sites have distances of less than 0.5°, with ρ and RMSE values of 0.89 and 2.6 μg/m^3, respectively. The ρ and RMSE for 92 sites (more than 0.5°) are 0.87 and 3.3 μg/m^3, respectively. The correlation coefficient between the RMSE and the distance is 0.33, and the correlation coefficient between the ρ and the distance is -0.17. The performance of the model decreases as the distance increases.

For Europe, 541 (64.8%) sites have distances of less than 0.5°, with ρ and RMSE values of 0.90 and 4.0 μg/m^3, respectively. The ρ and RMSE of the 293 sites (more than 0.5°) are 0.88 and 5.7 μg/m^3, respectively. The correlation coefficient between the RMSE and the distance is 0.19.

For China, 303 (19.5%) sites have a distance of less than 0.5°, with ρ and RMSE values of 0.95 and 9.5 μg/m^3, respectively. The ρ and RMSE for 1254 sites (more than 0.5°) are 0.91 and 12.1 μg/m^3, respectively. The correlation coefficient between the RMSE and the distance is 0.23. The correlation coefficient between ρ and distance is -0.71. As the distance increases, the correlation coefficient significantly decreases.

For India, the ρ and RMSE of 117 (56.8%) sites with a distance of less than 0.5° are 0.94 and 18.7 μg/m^3, respectively. The ρ and RMSE of 89 sites (more than 0.5°) are 0.89 and 18.8 μg/m^3, respectively. The correlation coefficient between ρ and distance is -0.36.

The above results indicate no significant correlation between model performance and distance in the United States and Europe, as these regions have adequate visibility stations. However, in China, India, and Canada, the performance of models is influenced by distance. Particularly in China, due to the limited number of visibility stations, although the correlation coefficient decreases with distance, there is no significant change in the RMSE. The correlation coefficient for visibility remains near 0.4. Even when the distance between two visibility stations reaches 1000 km, the maximum correlation coefficient for visibility remains near 0.4 (Fei et al., 2023). To acquire more $PM_{2.5}$ sample data, we do not disregard these distant sites since the models still shows a good performance for these sites. Nevertheless, more sufficient visibility stations in the same locations can enhance the model's performance.

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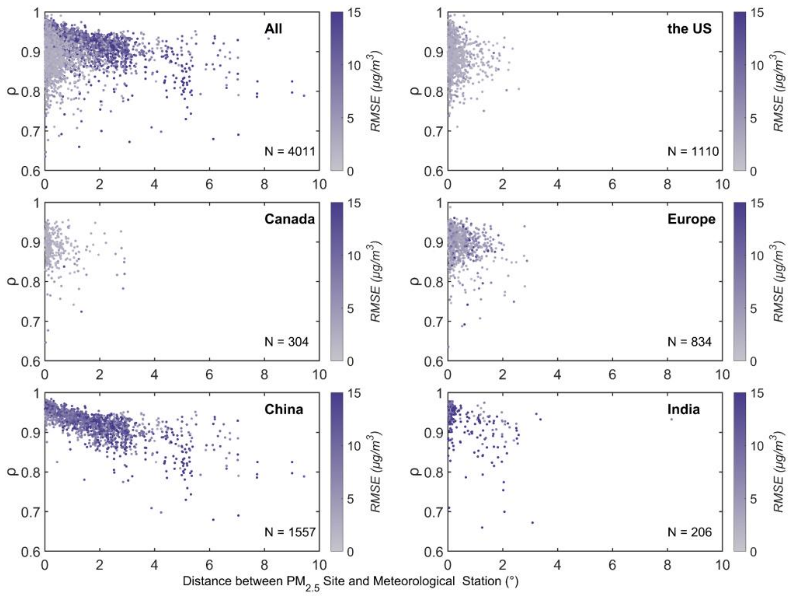

Figure 5 Scatter plots of the distance between the $PM_{2.5}$ site and visibility station and the model's correlation coefficient (ρ) for all sites and sites in the United States, Canada, Europe, China, and India. The color bar represents the root mean square error (RMSE) of the model. N is the number of sites.

3.3 Evaluation of Model's Predictive Ability

3.3.1 For All Data

A total of 1,149,152 pairs of test data is employed to evaluate the model's predictive ability. Figure 6 shows the density scatter plot between the predicted $PM_{2.5}$ concentration and the test $PM_{2.5}$ concentration. The results indicate that the linear regression coefficient is 0.862 ± 0.001 within a 95% confidence interval, R^2 is 0.80, RMSE is 13.5 μg/m^3, and MAE is 6.9 μg/m^3. Previous studies have shown that the R^2 range of the model's predictive results at the daily scale is 0.42-0.89, and the RMSE range is 9.59-32.09 μg/m^3 (Gui et al., 2020; Zhong et al., 2021). The test results exhibit excellent predictive capability.

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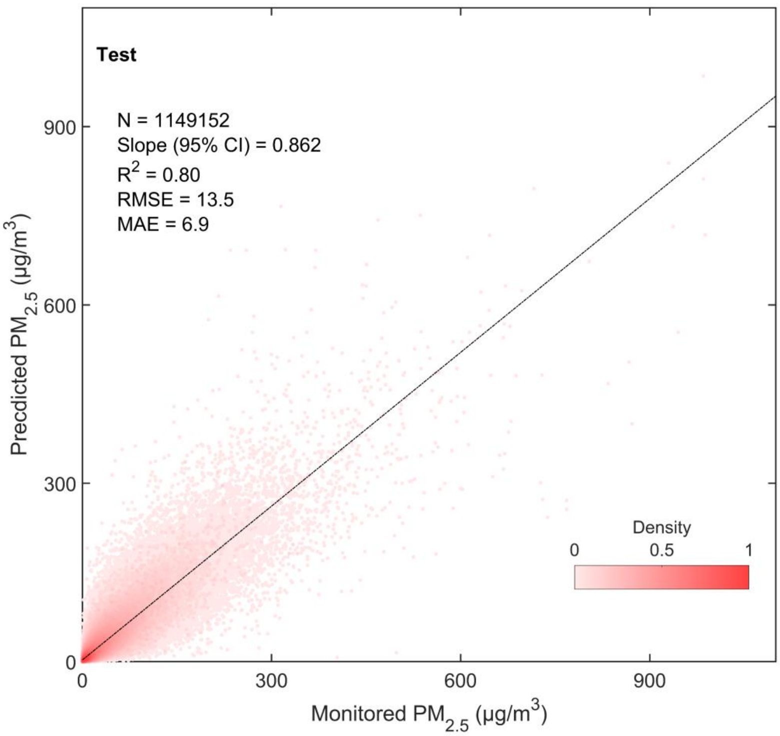

Figure 6 Density scatter plot (a) between the predicted $PM_{2.5}$ concentration and monitored $PM_{2.5}$ concentration of the test results at the daily scale. The dashed black line is the linear regression line. N is the length of the data pairs, and Slope is the linear regression coefficient within a 95% confidence interval (CI). R^2 is the coefficient of determination, RMSE is the root mean square error, and MAE is the mean absolute error.

3.3.2 For the Site and Region Scales

We analyze the test results for Canada, the United States, Europe, China, and India to assess the predictive ability of the model in different regions. Figure 7 shows the spatial distributions of the test RMSE, MAE, and ρ and their frequency and cumulative frequency distributions. Table 2 lists the test results of the metrics.

For all sites, the average RMSE is 12.60 μg/m^3. The RMSE-to-mean ratio is 48.6%. The average MAE is 8.52 μg/m^3. The MAE-to-mean ratio is 32.9%. The average ρ is 0.77.

For the United States, the RMSE, MAE, and ρ are 4.90 μg/m^3, 3.15 μg/m^3, and 0.71, respectively. For Canada, the RMSE, MAE, and ρ are 4.89 μg/m^3, 3.01 μg/m^3, and 0.74, respectively. The results in the United States and Canada are better in the west than in the east. The RMSE, MAE, and ρ for Europe are 7.54 μg/m^3, 4.91 μg/m^3, and 0.77, respectively. For China, the RMSE, MAE, and ρ are 20.16 μg/m^3, 13.81 μg/m^3, and 0.81, respectively. For India, the RMSE, MAE, and ρ are 28.84 μg/m^3, 19.57 μg/m^3, and 0.83, respectively. The results show that in developing regions (China and

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India), ρ is better than that in developed regions (the United States, Canada, and Europe), which means that the predictive ability of the model is better for severely polluted regions.

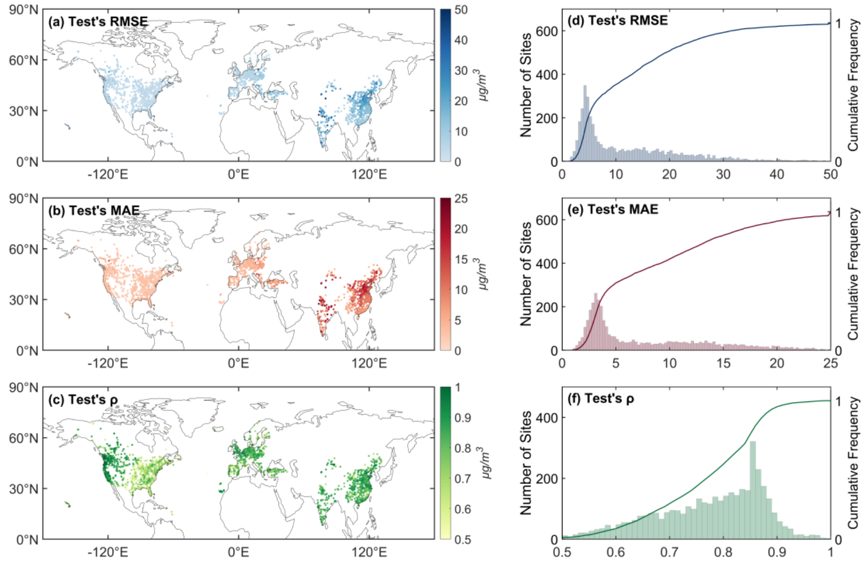

Figure 7 Spatial distribution (a-c) of the root mean squared error (RMSE), mean absolute error (MAE), and correlation coefficient (ρ) between the model's predicted values and test values. Number of sites (bar) and cumulative frequency (curve) (d-e) of the RMSE, MAE, and ρ.

Table 2 The test results of the model's performance metrics for all sites and sites in the United States, Canada, Europe, China and India.

Test	*RMSE ($\mu g/m^3$)*	*MAE ($\mu g/m^3$)*	*ρ (Pearson's correlation)*	*Mean ($\mu g/m^3$)*	*RMSE/Mean (%)*	*MAE/Mean (%)*
All	12.60	8.52	0.77	25.9	48.6	32.9
America	4.90	3.15	0.71	9.1	53.8	34.6
Canada	4.89	3.01	0.74	7.2	67.9	41.1
Europe	7.54	4.91	0.77	14.4	52.3	34.1
China	20.16	13.81	0.81	42.2	47.7	32.7
India	28.94	19.62	0.83	67.6	42.8	29.0

3.4 Uncertainties and Limitations

3.4.1 Uncertainty in the Pollution Level

Figure 8 shows the uncertainty in the predicted $PM_{2.5}$ concentration with respect to the pollution level of the monitored $PM_{2.5}$. For all sites, the uncertainty in the bias increases as the pollution level increases. The mean bias and the median bias shift from positive to negative with increasing pollution levels. The mean bias of 88.4% of the data is less than 2 $\mu g/m^3$. A mean bias of 86.9%

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(<40 μg/m^3) is positive, and a median bias of 38.9% (<8 μg/m^3) is positive. This result indicates that the model overestimates at low concentrations.

The bias for each region also increases with pollution level. For sites in the United States, the mean bias of 92.1% is less than 2 μg/m^3. A total of 69.1% (<10 μg/m^3) are positive. For sites in Canada, the mean bias of 82.5% is less than 2 μg/m^3. A total of 73.3% are positive (<8 μg/m^3). Among the data (<8 μg/m^3), 57.9% of the median is positive. For sites in Europe, the mean bias of 64.8% is less than 2 μg/m^3, and 69.8% is positive. A total of 49.0% of the median is positive. For sites in China, 81.8% of the bias is less than 5 μg/m^{3,} and 68.9% (<45 μg/m^3) is positive. A total of 48.0% (<30 μg/m^3) of the median is positive. For sites in India, 80.5% of the bias is less than 8 μg/m^3, and 73.5% (<80 μg/m^3) is positive. A total of 52.6% (<60 μg/m^3) of the median values are positive.

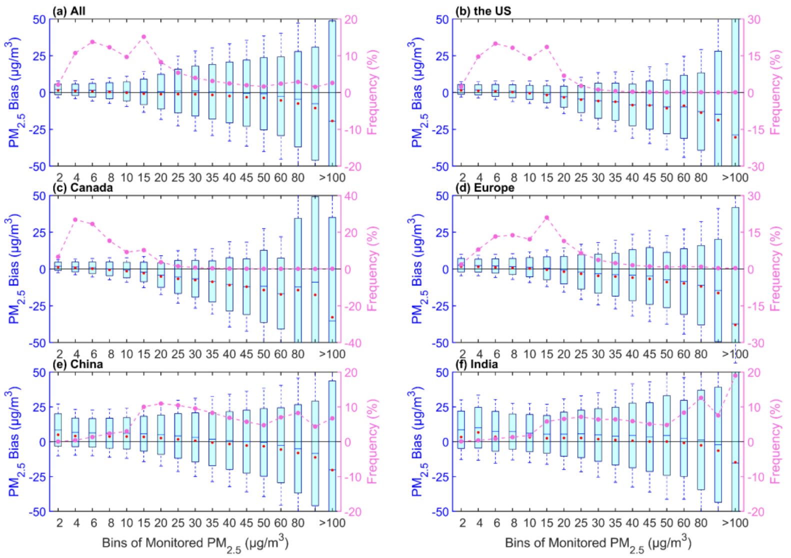

Figure 8 Boxplots of the pollution level and bias (predicted $PM_{2.5}$ - monitored $PM_{2.5}$) for all sites (a), sites in the United States (b), Canada (c), Europe (d), China (e), and India (f). The box's upper and lower limits represent ±1 standard deviation, the whiskers represent 2 times the standard deviation, the red circle represents the median, and the short line represents the mean bias. The frequency (%) on the right y-axis represents the percentage of data with different pollution levels (dashed line).

3.4.2 Uncertainty in the Station Elevation

With the spatial variability in $PM_{2.5}$, we analyze the mean bias at different station elevations. Figure 9 shows the relationships between the elevations of the visibility stations and the bias. The bias exhibits variations across different elevations for all sites. A total of 89.5% of the data are at an elevation of 1 km. The mean bias ranges from -0.1 to 0.5 μg/m^3. High uncertainties in bias occur at elevations below 0.2 km, 0.4-0.5 km, and 1-3 km. A total of 88.5% of the data have positive mean biases. Negative biases are observed at elevations of 0.6-0.8 km, 3 km, and 5 km. A total of 57.7% of the data have a positive median. This finding indicates a nonsignificant overestimation of the

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predicted $PM_{2.5}$ concentration due to the various elevations.

The bias patterns vary across regions. For the United States, 92.8% of the data correspond to elevations below 1 km. The mean bias ranges from -0.1 to 0.5 μg/m³. A total of 88.8% of the mean biases are positive, and the median of 99% is positive. For Canada, 90.1% of the data correspond to elevations below 1 km. The mean bias ranges from -0.1 to 0.2. A total of 46.5% of the mean bias is positive, and the median is positive except at elevations of 0.7 km and 4 km. A higher uncertainty in the bias occurs at elevations ranging from 0.5-0.8 km. For Europe, 92.9% of the data correspond to elevations below 1 km. The bias ranges from -0.2 to 0.2 μg/m³. A total of 62.7% of the mean bias is negative, and the median is positive. High standard deviations are observed at elevations of 0.2 km, 0.05 km, and 0.5-0.6 km. A significant bias occurs at 0.6 km. For China, 81.9% of the data correspond to elevations below 0.5 km. The median is positive, and the mean bias is positive except at 0.1 km. The lowest standard deviation occurs at an elevation of 0.3 km. For India, the mean bias ranges from -0.3 to 0.9 μg/m³. The highest bias occurs at an elevation of 0.3 km. There is a negative mean bias in the range of 0.1-0.4 km. The medians are positive except at an elevation of 0.4 km.

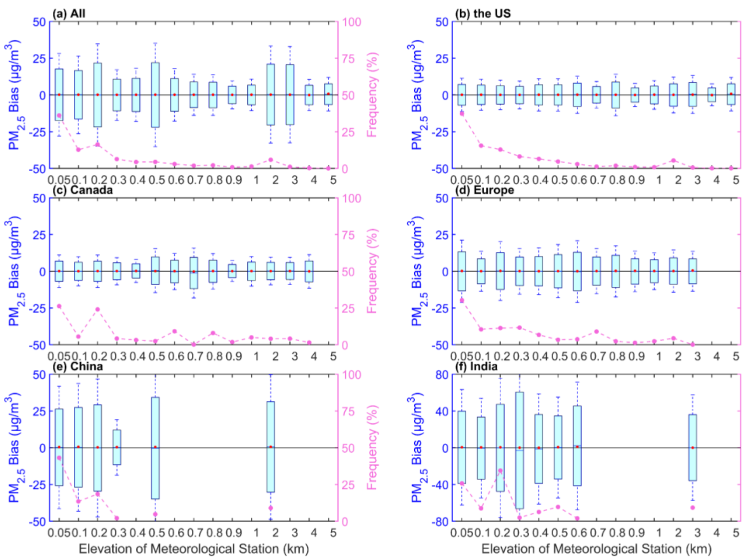

Figure 9 Boxplots of the elevation of the visibility station and bias (predicted $PM_{2.5}$ - monitored $PM_{2.5}$) for all sites (a), sites in the United States (b), Canada (c), Europe (d), China (e), and India (f). The box's upper and lower limits represent ±1 standard deviation, the whiskers represent 2 times the standard deviation, the red circle represents the median, and the short line represents the mean bias. The frequency (%) on the right y-axis represents the percentage of data at different pollution levels (dashed line).

3.4.3 Uncertainty in the Station Distance

As the visibility stations and $PM_{2.5}$ sites are not collocated, we analyze the $PM_{2.5}$ mean bias at different distances. Figure 10 shows the distance between the visibility of the station and the $PM_{2.5}$ site and bias. For all sites, the standard deviation gradually increases with distance, indicating an

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increase in uncertainty with increasing distance. Except at distances of 0.05° and 1°, the mean bias is positive. The median is positive. For each region, the distance of the largest average bias is 3° in the United States, 3° in Canada, 0.8° in Europe, 10° in China, and 0.4° in India. The distances are below 1° in the United States, Canada, Europe, and India, while they are 1-3° in China. This finding is due to the limited number of visibility sites in China. The mean bias exhibits greater uncertainties in China and India.

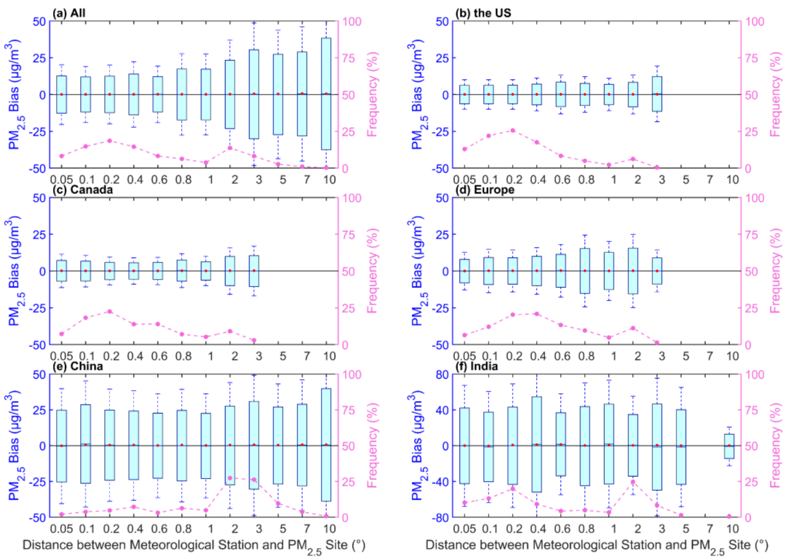

Figure 10 Boxplots of the distance between the visibility station and the $PM_{2.5}$ site and bias (predicted $PM_{2.5}$ - monitored $PM_{2.5}$) for all sites (a), sites in the United States (b), Canada (c), Europe (d), China (e), and India (f). The box's upper and lower limits represent ±1 standard deviation, the whiskers represent 2 times the standard deviation, the red circle represents the median, and the short line represents the mean bias. The frequency (%) on the right y-axis represents the percentage of data under different pollution levels (dashed line).

3.4.4 Discussion on the Uncertainties and Limitations

There are some uncertainties and limitations in this study. The upper limit of visibility ($PM_{2.5}$) is 10 km (1000 μg/m^3), which can cause some uncertainties in modeling. The maximum distance for spatial matching between the visibility stations and $PM_{2.5}$ monitoring sites is 10° due to the spatial variability in aerosols, which may increase the uncertainty in the estimated $PM_{2.5}$ concentration. The boundary layer height is closely related to the vertical structure of $PM_{2.5}$, and reanalysis data may introduce uncertainty to the model. Because of the nonuniform vertical distribution of aerosols, the different elevations of the visibility stations and the $PM_{2.5}$ monitoring sites further increase the uncertainty in estimating $PM_{2.5}$. In addition, the spatial coverage of visibility stations, especially in China, is limited, which may increase the uncertainty in the representativeness of regional $PM_{2.5}$ trends and pollution levels. With the increasing human concern about air pollution and the implementation of pollution control measures, the types of major atmospheric pollutants have

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changed, the composition of particulate matter has also evolved, the scattering and absorption characteristics may have changed, and the relationship between visibility and $PM_{2.5}$ may change. These changes may lead to uncertainty in estimating historical $PM_{2.5}$, especially before 2000 (ground and satellite observations are limited). Despite these limitations, we establish a long-term $PM_{2.5}$ dataset based on visibility from 1959 to 2022, providing insights into the long-term spatiotemporal characteristics of $PM_{2.5}$ in the Northern Hemisphere.

4 Comparisons with Other $PM_{2.5}$ Datasets

We compare the monthly estimated $PM_{2.5}$ with the $PM_{2.5}$ of those derived from a satellite AOD and two reanalysis datasets, including (1) ACAG, the monthly satellite-derived $PM_{2.5}$ from 1998 to 2022 (Van Donkelaar et al., 2019; Hammer et al., 2020); (2) MERRA-2, the monthly $PM_{2.5}$ from 1980 to 2022 (Buchard et al., 2016; Buchard et al., 2017; Gelaro et al., 2017); and (3) CAMS, the monthly $PM_{2.5}$ from 2003 to 2022 (Inness et al., 2019). The time ranges for comparing the estimated $PM_{2.5}$ with the ACAG, MERRA-2, and CAMS data are 1998-2022, 1980-2022, and 2003-2022, respectively. The monthly average should meet a minimum requirement of at least ten days per month.

4.1 Monthly Frequency and Annual Cycle of $PM_{2.5}$

We compare the frequency of the estimated $PM_{2.5}$ concentration at different pollution levels, with an interval of 1 $\mu g/m^3$, with three other datasets. Figure 11 shows the monthly $PM_{2.5}$ frequencies of the estimated, ACAG, MERRA-2, and CAMS datasets for all sites and regional sites.

Compared with the ACAG data, they exhibit similar frequency distributions. However, the frequency of estimated $PM_{2.5}$ concentrations is greater at high pollution levels at all sites. Regionally, the frequency distributions are similar at different pollution levels in the United States and Canada. In Europe, China, and India, the frequency of high concentrations is greater than that of the ACAG.

Compared with the MERRA-2 data, the frequency distribution of the estimated data is similar to that of the ACAG for all the sites. Regionally, the frequency distributions of the estimates are comparable in the United States and Canada. However, in Europe, China, and India, the differences in the frequency of high pollution levels are greater than those in the ACAG.

Compared with the CAMS data, the frequency distributions at high pollution levels are similar, but the frequency at high pollution levels is lower. Regionally, Europe differs from other regions, as the frequency of high pollution levels is higher.

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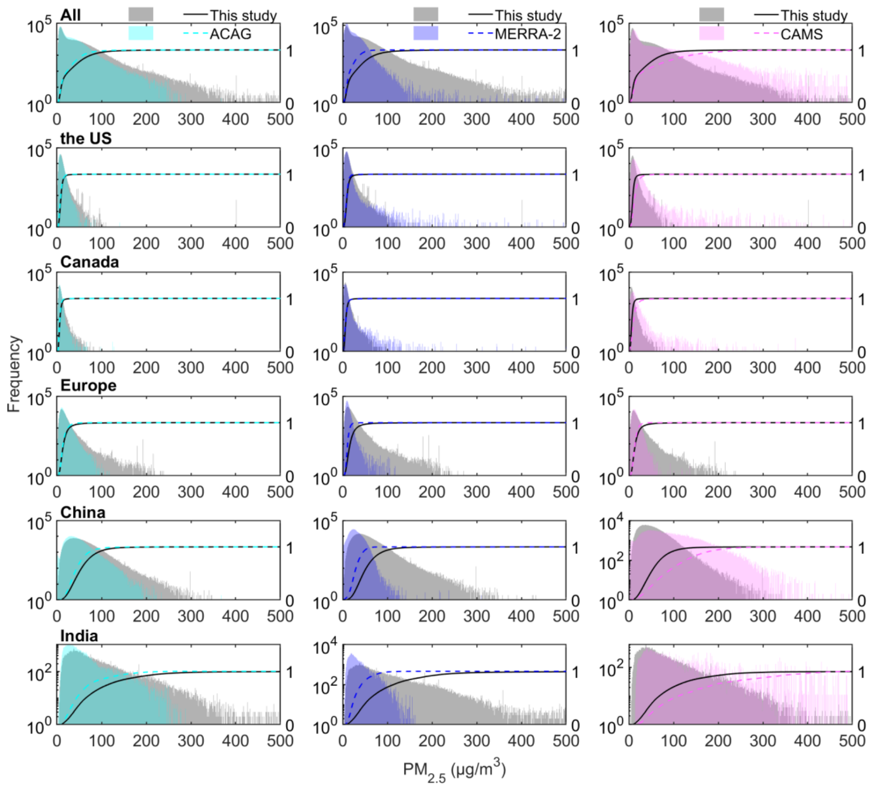

Figure 11 Frequency (left axis) and cumulative frequency (right axis) of monthly $PM_{2.5}$. The time range of the estimated $PM_{2.5}$ corresponds to the time range of the three datasets (ACAG from 1998 to 2022, MERRA-2 from 1980 to 2022, and CAMS from 2003 to 2022). The bins range from 0 to 500 μg/m^3 with an interval of 1 μg/m^3.

In Figure 12, we compare the multiyear monthly average $PM_{2.5}$ concentration with that of the three datasets. For all sites, the correlation coefficients between the estimated and ACAG, MERRA-2, and CAMS data are 0.99, 0.42, and 0.93, respectively, and the average biases (average relative biases) are 6.6 μg/m^3 (26%), 14.1 μg/m^3 (76%), and -19.1 μg/m^3 (-37%), respectively. The estimated multiyear average monthly $PM_{2.5}$ concentrations are higher for ACAG and MERRA-2 and lower for CAMS. The correlation coefficient is highest for ACAG and lowest for MERRA-2.

Compared with the ACAG data, the correlation coefficients are 0.97, 0.96, 0.98, 0.99, and 0.99, with average biases (average relative biases) of 0.8 μg/m^3 (9%), 0.5 μg/m^3 (7%), 2.2 μg/m^3 (16%), 10.8 μg/m^3 (26%), and 31.4 μg/m^3 (62%) in the United States, Canada, Europe, China, and India, respectively. The annual variations in the two datasets are nearly consistent across all regions. The bias is less than 10% for the United States and Canada, while India exhibits the largest bias.

Compared with the MERRA-2 data, the correlation coefficients are 0.30, 0.61, -0.25, 0.80, and 0.45, with average biases (average relative biases) of 1.1 μg/m^3 (16%), 0.2 μg/m^3 (5%), 7.5 μg/m^3 (67%), 24.1 μg/m^3 (83%), and 56.1 μg/m^3 (169%) in the United States, Canada, Europe, China, and India,

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respectively. There are differences in the annual variations between the two datasets, particularly during winter (November to January) and spring (February to March), in all regions. The largest difference occurs in March and September to December in Europe, showing the opposite trend. The highest correlation coefficient is observed in China, which has the second largest bias. The largest bias is in India.

Compared with the CAMS data, the correlation coefficients are 0.29, 0.22, 0.02, 0.91, and 0.98, with average biases (average relative biases) of -5.4 μg/m³ (-34%), -5.0 μg/m³ (-38%), 2.7 μg/m³ (21%), -38.7 μg/m³ (-42%), and -52.7 μg/m³ (-36%) in the United States, Canada, Europe, China, and India, respectively. The annual variations between the CAMS and ACAG data are similar in China and India but have more significant biases. The smallest differences in the United States and Canada occur in January and December. In Europe, the months with more significant biases are January to March and September to December, while biases are smaller in other months.

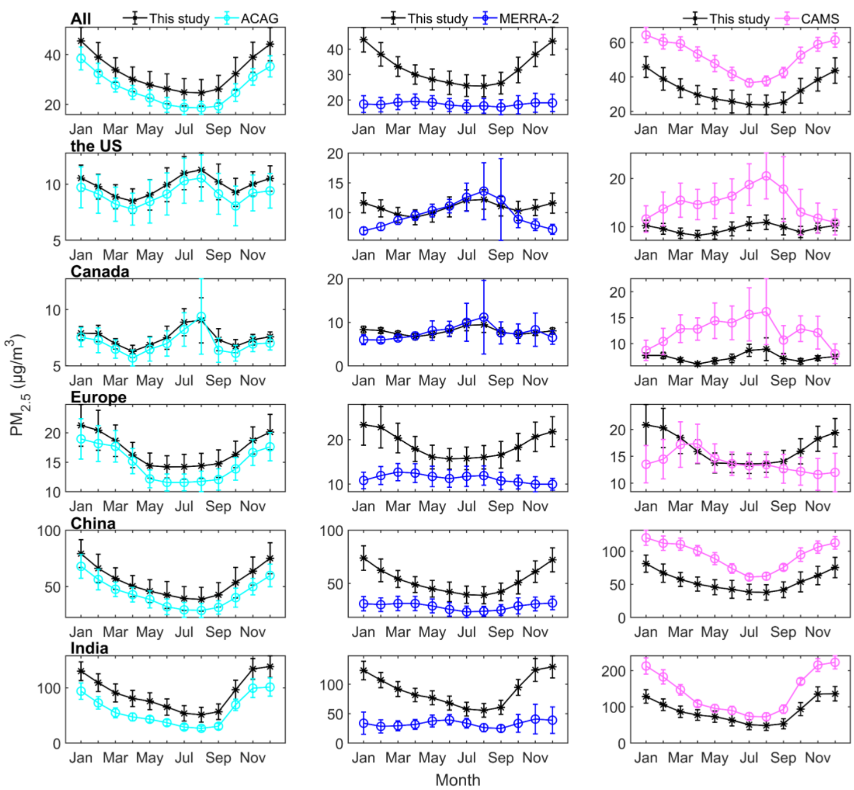

Figure 12 Multiyear monthly average $PM_{2.5}$ of our data and the three datasets. The time range of the estimated $PM_{2.5}$ corresponds to the time range of the three datasets (ACAG data from 1998 to 2022, MERRA-2 data from 1980 to 2022, and CAMS data from 2003 to 2022).

4.2 Time Series at the Annual Scale

Figure 13 shows the annual average $PM_{2.5}$ concentration from 1959 to 2022 in different regions, along with a comparison to the $PM_{2.5}$ concentrations derived from other datasets. Another dataset is

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used for comparison in China: the monthly $PM_{2.5}$ of the CHAP from 2000 to 2021 (Wei et al., 2020b; Wei et al., 2021). We use correlation coefficients, mean bias and mean relative bias to compare the relationships and differences among the $PM_{2.5}$ datasets.

In the United States, the estimated $PM_{2.5}$ concentrations exhibit correlation coefficients of 0.96, 0.88, and -0.38 with the ACAG, CAMS, and MERRA-2 data, respectively; the mean bias (mean relative bias) is 0.8 (10%), -5.4 (-35%), and 1.1 (13%) for each dataset, respectively.

In Canada, the estimated $PM_{2.5}$ concentrations exhibit correlation coefficients of 0.84, 0.62, and -0.46 with the ACAG, CAMS, and MERRA-2 data, respectively; the mean bias (mean relative bias) is 0.5 μg/m^3 (7%), -5.1 μg/m^3 (-40%), and 0.2 μg/m^3 (6%) for each dataset, respectively.

In Europe, the estimated $PM_{2.5}$ concentrations exhibit correlation coefficients of 0.96, 0.96, and 0.76 with the ACAG, CAMS, and MERRA-2 data, respectively; the mean bias (mean relative bias) is 2.3 μg/m^3 (15%), 2.6 μg/m^3 (20%), and 7.5 μg/m^3 (66%) for each dataset, respectively.

In China, the estimated $PM_{2.5}$ concentrations exhibit correlation coefficients of 0.78, 0.98, 0.81, and 0.51 with the ACAG, CHAP, CAMS, and MERRA-2 data, respectively; the mean bias (mean relative bias) is 10.7 μg/m^3 (24%), 2.5 μg/m^3 (4%), -39.1 μg/m^3 (-42%), and 24 μg/m^3 (90%) for each dataset, respectively.

In India, the estimated $PM_{2.5}$ concentrations exhibit correlation coefficients of -0.3, -0.02, and -0.09 with the ACAG, CAMS, and MERRA-2 data, respectively; the mean bias (mean relative bias) is 29.9 μg/m^3 (53%), -58.9 μg/m^3 (-40%), and 56.1 μg/m^3 (203%) for each dataset, respectively. From 2013 to 2022, the correlation coefficients with the ACAG and CAMS data are 0.71 and 0.70, respectively. The trend of visibility declines from 1961 to 2008. The frequency of visibility (exceeding 10 km) in the afternoon decreases by 46%, and the frequency of visibility (below 4 km) in the morning increases by 21% (Jaswal et al., 2013), particularly in the central and northern regions. The low cloud cover significantly increases from 1960 to 2010 in the Indo-Gangetic Plain and the northwestern and eastern coasts of India (Jaswal et al., 2017). The average total cloud cover is 3.4 okta from 1960 to 2007, with a decrease of 0.07 okta/decade (Jaswal, 2010). However, the indirect impact of aerosols on cloud formation do not influence cloud cover (Ramanathan et al., 2005). The prevalence of clouds poses challenges for satellite retrievals in these areas, potentially contributing to substantial disparities between $PM_{2.5}$ concentrations estimated based on visibility and satellite retrievals. The CAMS reanalysis data are calibrated using satellite data and thus show consistency with the trend in AOD retrievals from satellites; the anthropogenic emission data are from the MACCity inventory (Inness et al., 2019), and there are significant variations among different anthropogenic emission inventories, particularly before 2010, which leads to substantial uncertainties in India (Granier et al., 2011; Liu et al., 2022). These issues exist to a greater or lesser extent in other regions, which may contribute to the increased disparities between estimated $PM_{2.5}$ and reanalysis data before 2012.

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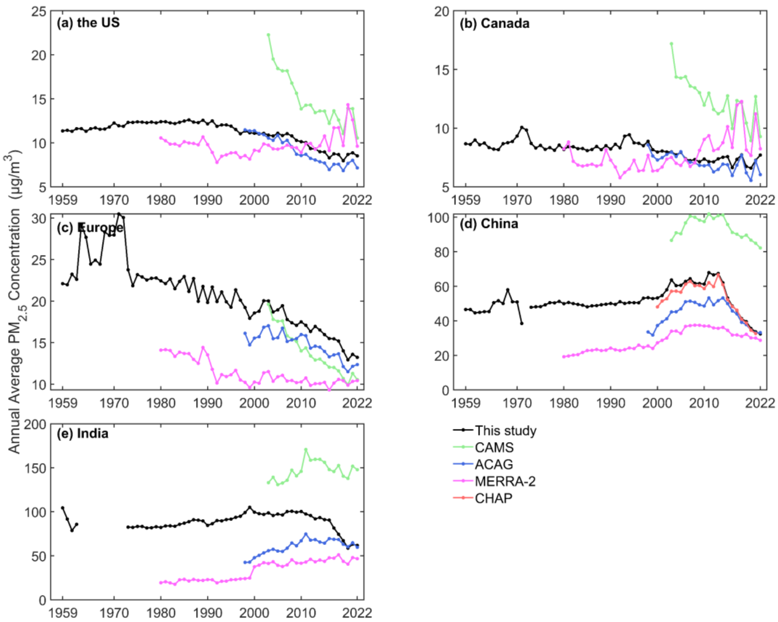

Figure 13 Annual mean $PM_{2.5}$ concentration from 1959 to 2022 in the United States (US) (a), Canada (b), Europe (c), China (d), and India (e). The other four datasets are ACAG from 1998 to 2022, CHAP from 2000 to 2021, MERRA-2 from 1980 to 2022, and CAMS from 2003 to 2022.

4.3 Discussion on the Differences among the $PM_{2.5}$ Datasets

$PM_{2.5}$ is considered a pollutant that decreases visibility. There is a negative correlation between visibility and $PM_{2.5}$ concentration, and the reciprocal of visibility is proportional to the extinction coefficient, which is closely related to the concentration of particulate matter (Wang et al., 2012; Zhang et al., 2017; Zhang et al., 2020). Prior to the widespread implementation of $PM_{2.5}$ measurements or lack of measurement of particulate matter, visibility is often used as a proxy for particulate matter pollution (Huang et al., 2009; Singh et al., 2020). It is the basis for using visibility to estimate $PM_{2.5}$ concentration. Studies have shown that meteorological observations such as temperature and humidity also play an important role in estimating $PM_{2.5}$ concentration using visibility (Shen et al., 2016; Xue et al., 2019; Zhong et al., 2021). The advantages of ground-based visibility and other meteorological variables observations include long-term records, high temporal resolution, and good data completeness, and the visibility observations from airports can be traced back to 1959 in this study. Therefore, we employ a machine learning approach to establish the relationship between $PM_{2.5}$ and visibility and other meteorological variables, and estimate the long-term historical $PM_{2.5}$ concentration from 1959 to 2022, and discuss the limitations and uncertainties. It should be noted that not all sites of $PM_{2.5}$ have the time range from 1959 to 2022, which depends on the record length of matched visibility station.

There are differences between $PM_{2.5}$ based on visibility, $PM_{2.5}$ based on satellite retrievals, and

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$PM_{2.5}$ of reanalysis. $PM_{2.5}$ based on satellite retrievals typically requires consideration of aerosol vertical profiles usually provided by observations, assumptions, or chemical transport models to obtain the aerosol properties near the surface (Van Donkelaar et al., 2010; Wei et al., 2019b; Van Donkelaar et al., 2021). $PM_{2.5}$ from reanalysis usually requires accurate meteorological fields and emission inventories. Although ERA5 has provided meteorological reanalysis since 1940, the historical emission inventories and physical-chemical mechanisms in the chemical transport model still have significant uncertainties, which increase the uncertainty in particulate matter concentration. Additionally, the assimilated data in reanalysis mainly consist of satellite AOD and ground-based AOD, aiming to improve column aerosol properties, without considering near-surface $PM_{2.5}$ (Buchard et al., 2017; Gelaro et al., 2017; Provençal et al., 2017; Huijnen et al., 2019; Inness et al., 2019; Ali et al., 2022). These factors contribute to the differences in estimating $PM_{2.5}$ concentration among the three methods.

In this study, the upper limit of the estimated daily $PM_{2.5}$ concentration is set to 1000 μg/m^3 because the $PM_{2.5}$ concentration is greater than 500 during heavy pollution weather, which may contribute to the higher frequency at high pollution levels than in the other datasets. We do not delete visibility records during sand and dust weather when preprocessing the data, which may lead to an overestimation of $PM_{2.5}$ in dusty areas, such as northern China and northwestern India.

The frequency and monthly/annual variations in our data are consistent with those of $PM_{2.5}$ based on satellite retrievals (ACAG and CHAP). The concentration level is higher than in those datasets because their upper limits are lower. The AOD is a physical quantity that describes the properties of aerosol columns. It is important to consider the vertical structure of aerosols when establishing a connection between AOD and near-ground $PM_{2.5}$. Van Donkelaar et al. (2006; 2010) demonstrated that aerosol vertical profile errors in chemical transport models and AOD retrieval and sampling result in an approximately 25% uncertainty of one standard deviation. Sensitivity testing shows that a 1% estimation error in the AOD can lead to a 0.27% estimation error in the $PM_{2.5}$ concentration (Wei et al., 2021). Visibility is a near-surface observation that is not affected by clouds or surface types and has high temporal resolution (Liu et al., 2017; Singh et al., 2020; Zhong et al., 2021). In section 3.4, the uncertainty analysis provides an explanation for the overestimation.

In section 2.6.3, we introduce the chemical model, emission, and assimilation of MERRA-2. The $PM_{2.5}$ concentration from MERRA-2 does not include nitrates, and the assimilation of AOD mainly provides constraints on aerosols after 2000 (Buchard et al., 2016; Randles et al., 2017; Ali et al., 2022). The lack of nitrate is a limitation in areas with high nitrate concentrations. For example, an extreme pollution event over China in January 2013 is not captured well (Buchard et al., 2017). Ali et al. (2022) used 1.4 × [SO_4^{2-}] to represent nitrate concentration, and the results showed a correlation coefficient of 0.55 with the observed $PM_{2.5}$. Compared to the ACAG over the United States, which has a low nitrate concentration, the MERRA-2 surface $PM_{2.5}$ concentration is greater in rural areas than in urban and suburban areas, with high and localized emissions reducing the representation of the grid mean $PM_{2.5}$ (Buchard et al., 2017). Therefore, the lack of nitrate and insufficient assimilation data are the key factors leading to the significant differences between the two datasets.

In section 2.6.4, we introduce the CAMS $PM_{2.5}$. The $PM_{2.5}$ concentration from CAMS is significantly greater than the estimated $PM_{2.5}$ concentration and follows a similar annual cycle,

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except in Europe. In Europe, the CO and NO_2 concentrations in CAMS are lower than those in winter (Flemming et al., 2015), which may lead to the underestimation of nitrate emissions and its precursors, resulting in the underestimation of $PM_{2.5}$ concentrations. Some studies have reported similar results (Kong et al., 2021; Ryu and Min, 2021; Ali et al., 2022; Jin et al., 2022). This finding may be related to the vertical section structure, composition, and microphysical properties of aerosols (Ali et al., 2022). Because NO_2 emissions are obtained by multiplying CO emissions by a factor of 0.2, the uncertainty in nitrate increases. Studies have shown that the uncertainties in MACCity (Huijnen et al., 2019) and dust (Ukhov et al., 2020) also cause overestimation in CAMS $PM_{2.5}$.

Overall, our $PM_{2.5}$ dataset has good consistency with $PM_{2.5}$ based on satellite AOD data. There are some differences in the reanalysis $PM_{2.5}$ concentrations. We also hope that our dataset can provide auxiliary support for reanalysis datasets.

5 $PM_{2.5}$ Variability from 1959 to 2022

5.1 Monthly $PM_{2.5}$ and Trend

Figure 14 (a) shows the frequency of the estimated monthly $PM_{2.5}$ from 1959 to 2022, and Table 3 lists the maximum frequency for each region. The order of the concentrations with the greatest frequency was Canada (8 μg/m^3), the United States (12 μg/m^3), Europe (18 μg/m^3), China (42 μg/m^3) and India (64 μg/m^3). Canada and the United States are areas with less frequent $PM_{2.5}$ pollution. $PM_{2.5}$ pollution occurs frequently in China and India. The results indicate that the $PM_{2.5}$ concentrations in developed countries are significantly lower than those in developing countries in the Northern Hemisphere.

Figure 14 (b-f) shows the anomalies of the estimated monthly $PM_{2.5}$ concentration from 1959 to 2022, and Table 3 lists the trends for each region. The trends in each region from 1959 to 2022 are all negative; however, the trend in India does not pass the significance test ($p>0.05$). The fastest downward trend is in Europe, at -1.93 μg/m^3/decade. The trends in different regions vary at different times. Positive trends are detected in the United States from 1959 to 1990, in Canada from 1959 to 1993, and in China and India from 1959 to 2012. The most rapid upward trend is observed in India, at 3.35 μg/m^3/decade from 1959 to 2012. Negative trends are detected in the United States from 1991 to 2022, in Europe from 1959 to 1972 and from 1973 to 2022, and in China and India from 2013 to 2022. The most significant downward trend is observed in India, at -42.84 μg/m^3/decade. These regional trends are similar to those of previous studies in different periods (Van Donkelaar et al., 2010; Wang et al., 2012; Boys et al., 2014; Ma et al., 2016; Li et al., 2017; Hammer et al., 2020).

The trends in $PM_{2.5}$ concentration changes in different regions are closely associated with the implementation of relevant policies. The earlier pollution control measures are taken, the earlier the decreasing trend in the $PM_{2.5}$ concentration occurs, and the lower the threat of particulate matter pollution is to humans. In 1997, the United States EPA classified $PM_{2.5}$ as a hazardous substance in the National Ambient Air Quality Standard, and subsequent regulations in 2006 further strengthened the source control and management of fine particulate matter (Gilliam and Hall, 2016). In 1988, the Canadian federal government enacted the Canadian Environmental Protection Act, which enhanced the regulation of $PM_{2.5}$ (Davies, 1988). The European Union introduced the Air Quality Directive in 1996, followed by multiple revisions and updates to regulate and restrict air

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pollutants, including $PM_{2.5}$ (Kuklinska et al., 2015). However, Europe stands out due to its early adoption of clean production practices in heavy industries since the 1970s. Since 2012, China has implemented numerous regulations and standards for $PM_{2.5}$. For instance, the Monitoring Method for Atmospheric Particulate Matter ($PM_{2.5}$) was issued in 2012, and the Chinese Ministry of Environmental Protection released the Ambient Air Quality Standards in 2013, which include emission standards for $PM_{2.5}$ (Zhao et al., 2016a). In 2009, the Indian Ministry of Environment and Forests issued the National Ambient Air Quality Standards, which include control standards for air pollutants, including $PM_{2.5}$. Since 2015, the Indian government launched the National Clean Air Programme (NCAP) to improve air quality in India by implementing a series of measures to reduce the emissions of $PM_{2.5}$ and other pollutants (Ganguly et al., 2020). These environmental regulations have contributed significantly to the decline in $PM_{2.5}$ concentrations.

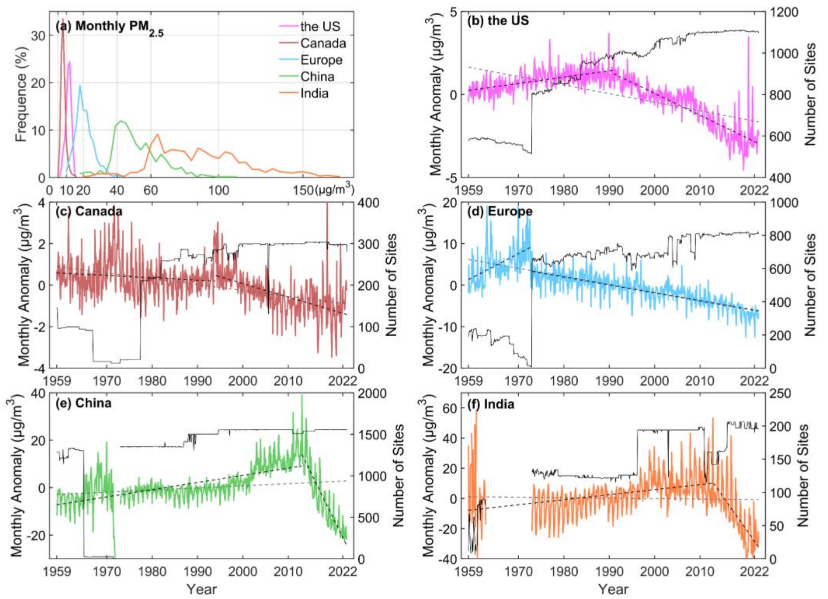

Figure 14 Frequency (a) and anomalies (b-f) of monthly $PM_{2.5}$ from 1959 to 2022 in the United States (the US), Canada, Europe, China, and India. The right Y-axis (b-f) is the monthly number of sites.

Table 3 The frequency and trend of the monthly $PM_{2.5}$ concentration from 1959 to 2022 in the United States (the US), Canada, Europe, China and India.

	Concentration Mode (μg/m³) and maximum frequency (%)	*Trend (μg/m³/decade)*		
the US	12 (24.3%)	-0.52* (1959-2022)	0.38* (1959-1990)	-1.32* (1991-2022)
Canada	8 (33.5%)	-0.28* (1959-2022)	-0.11* (1959-1993)	-6.48* (1994-2022）
Europe	18 (19.4%)	-1.93* (1959-2022)	5.69* (1959-1972)	-1.91* (1973-2022)
China	42 (11.9%)	-0.89*	3.04*	-38.82*

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		(1959-2022)	(1959-2012)	(2013-2022)
India	64 (9.1%)	-0.31 (1959-2022)	3.35* (1959-2012)	-42.84* (2013-2022)

The symbol * indicates passing the significance test, $p<0.01$; otherwise, not passing the significance test, $p>0.05$.

5.2 Annual $PM_{2.5}$ and Distribution

We analyze the spatial distribution of the multiyear average $PM_{2.5}$ concentration in each region, and we investigate the yearly variations in the spatial distribution based on the SDE and the average center, as shown in Figure 15. The mean center and SDE describe the periodic changes in the spatial distribution and dispersion of the $PM_{2.5}$ concentration in each region. The larger the ellipse area is, the more dispersed the spatial distribution of $PM_{2.5}$ is. The flatter the ellipse is, the stronger the spatial correlation of $PM_{2.5}$, and the direction of the major axis indicates the direction of the concentration.

The multiyear average $PM_{2.5}$ concentrations from 1959 to 2022 are 11.2 μg/m^3 in the United States, 8.2 μg/m^3 in Canada, 20.1 μg/m^3 in Europe, 51.3 μg/m in China, and 88.6 μg/m^3 in India. $PM_{2.5}$ concentrations in developed regions (North America and Europe) are significantly lower than those in developing regions (China and India).

For the United States, the concentration in the eastern region is greater than that in the western region. The $PM_{2.5}$ concentration at most sites in the eastern region is greater than 10 μg/m^3. Based on the area of the SDE, the spatial distribution is divided into three stages: 1959-1972, 1973-1976, and 1977-2022. The area decreases and then increases, indicating a changing trend in the spatial extent of the $PM_{2.5}$ concentration. The concentration distribution direction is east–west and rotates northward, and the mean center gradually moves northwest after 1977, indicating an increase in the $PM_{2.5}$ contribution in the western region.

For Canada, the concentrations in the eastern and western regions are greater than those in the central region. The area of the ellipse increases and then decreases. The concentration distribution direction is northwest-to-southeast, and the concentration rotates southward after 1977, indicating an increase in weight in the western region. The mean center gradually moves northwestward and then southeastward.

For Europe, high-concentration areas are mainly located in the central and eastern regions. The ellipse's area can be divided into three stages: 1959-1967, 1968-1972, and 1973-2022. The spatial variability decreases and then increases, corresponding to the mean centers moving north, south, and north. The concentration direction is northwest–southeast, and the major axis shortens after 1993, indicating that the directionality of the concentration weakens.

For China, high-concentration areas are in the central and eastern regions. The center of the SDE is located in the northeast region from 1965 to 1971, which may be related to Northeast China being the center of heavy industry during that period. After 1988, the area of the SDE increases significantly, and the center moves significantly southwestward and gradually northward after 2008. This finding indicates that the spatial distribution of $PM_{2.5}$ increases in a discrete pattern after 1988, and the concentration weight in the eastern region gradually increases. After 2008, the weight in the western region decreases again.

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For India, the highest concentration is in the northern region, and the lowest concentration is in the southern region. The area, shape, and mean center of the SDE show significant changes and can be divided into three stages. The SDE flattens between 1959 and 1962. The flattening weakens, and the area increases from 1963 to 1995. The spatial variability in $PM_{2.5}$ increases, and the mean center moves southward. From 1996 to 2022, the flattening further weakens, the area decreases, the spatial variability in $PM_{2.5}$ decreases, and the mean center shifts northward.

Above all, the concentration distributions in the United States and India exhibit an east–west pattern. The concentration distribution in Canada and Europe shows a northwest-to-southeast concentration gradient. In China, the $PM_{2.5}$ concentration distribution ranges from northeast to southwest. There are strong correlations between the $PM_{2.5}$ concentration and the location of the sites in Europe and Canada. However, the spatial correlation in India is gradually weakening, and the spatial dispersion of $PM_{2.5}$ in China is increasing. Studies have shown that the variation in $PM_{2.5}$ based on the mean center and the SDE is related to several factors, such as the energy structure, urbanization process, population distribution and vegetation coverage (Shi et al., 2018; Wu et al., 2018; Li et al., 2019; Wang et al., 2019; Lim et al., 2020; Qi et al., 2023).

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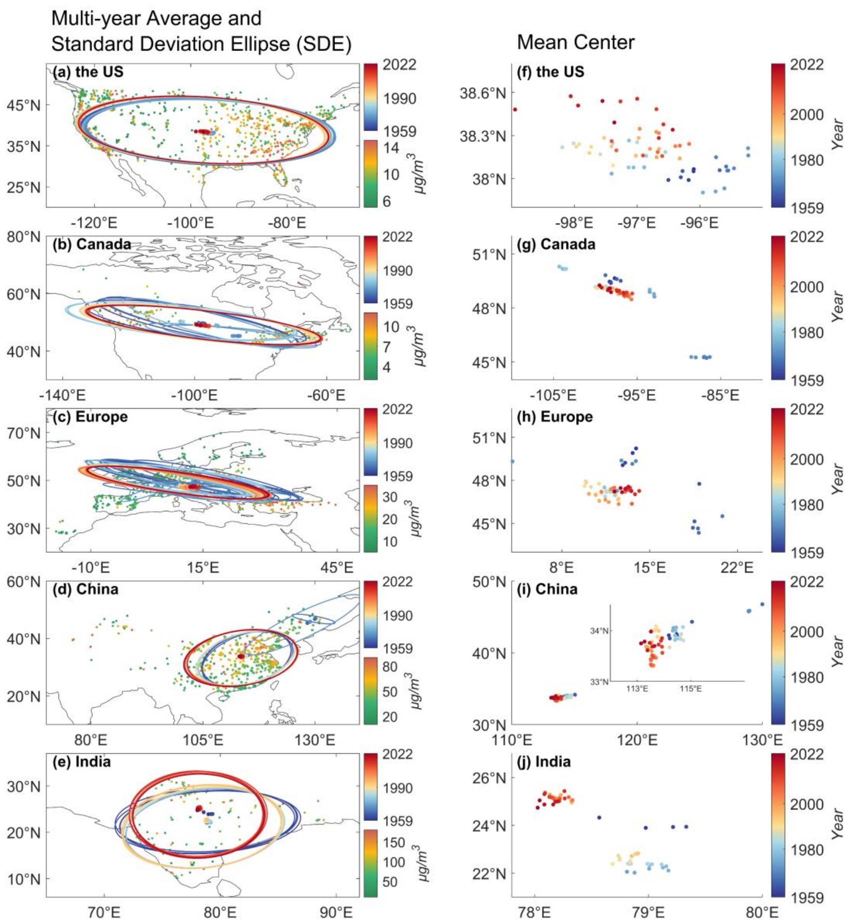

Figure 15 The spatial distribution of the multiyear average and standard deviation ellipse (SDE) (a-e) and the mean center (f-j) of the $PM_{2.5}$ concentration from 1959 to 2022 in the United States (the US), Canada, Europe, China, and India. The mean center and SDE describe the changes in the spatial distribution. The larger the ellipse area is, the more dispersed the spatial distribution of $PM_{2.5}$ is. The flatter the ellipse is, the stronger the spatial correlation of $PM_{2.5}$ is. The direction of the major axis indicates the direction of the concentration.

6 Conclusions

This study uses a machine learning method to estimate daily $PM_{2.5}$ for 4011 terrestrial sites in the Northern Hemisphere from 1959 to 2022 based on hourly visibility and related meteorological variables. Eighty percent of the sample data are used to train the model, and 20% are used for testing. The model's performance and predictive ability are evaluated. We analyze the uncertainty and discuss the limitations of the dataset. We compare the estimated $PM_{2.5}$ with the $PM_{2.5}$ based on the satellite AOD and $PM_{2.5}$ of the reanalysis datasets. Finally, the $PM_{2.5}$ variability in each region over the past 64 years is analyzed. We hope our dataset will be useful for studying the atmospheric environment, human health, and climate change and provide auxiliary support for assimilation. Several key results of this study are described as follows:

The most important variable Visibility is the most important variable at 79.1% of the $PM_{2.5}$ sites, as visibility can also be considered an indicator of $PM_{2.5}$ without fog or precipitation. Other meteorological variables play a secondary role in the model, especially temperature and dew point temperature. Visibility can serve as a good indicator of $PM_{2.5}$.

Model performance and predictive ability. The training results show that the slope between the estimated $PM_{2.5}$ concentration and the monitored $PM_{2.5}$ concentration within the 95% confidence interval is 0.946, the R^2 is 0.95, the RMSE is 7.0 μg/m³, and the MAE is 3.1 μg/m³. The test results show that the slope between the predicted $PM_{2.5}$ concentration and the monitored $PM_{2.5}$ concentration is 0.862 ± 0.0010 within a 95% confidence interval, R^2 is 0.80, RMSE is 13.5 μg/m³, and MAE is 6.9 μg/m³. The model shows good stability and predictive ability.

Comparison with other datasets. The estimated $PM_{2.5}$ concentration is consistent with the $PM_{2.5}$ concentration based on satellite AOD data at the monthly scale. The correlation coefficient of the annual cycles in each region is greater than 0.96. Compared with the reanalysis data, there are some differences among regions, which are closely related to the accuracy of emission inventories and the vertical structures of aerosols.

Monthly $PM_{2.5}$. From 1959 to 2022, the $PM_{2.5}$ concentration at the highest frequency is 12 μg/m³, 8 μg/m³, 17 μg/m³, 40 μg/m³ and 63 μg/m³, and the trends are -0.52 μg/m³/decade, -0.28 μg/m³/decade, -1.93 μg/m³/decade, -0.89 μg/m³/decade, and -0.31 μg/m³/decade, respectively, for the United States, Canada, Europe, China, and India. $PM_{2.5}$ concentrations in all regions show a periodic increase and decrease from 1959 to 2022. The decreasing trends are -1.32 μg/m³/decade from 1991 to 2022 in the United States, -6.48 μg/m³/decade from 1994 to 2022 in Canada, -1.91 μg/m³/decade from 1973 to 2022 in Europe, and -38.82 μg/m³/decade and -42.84 μg/m³/decade from 2013 to 2022 in China and India, respectively. Although the $PM_{2.5}$ concentrations in developing countries are significantly greater than those in developed countries, they have declined more quickly in recent years.

Annual $PM_{2.5}$. The multiyear average $PM_{2.5}$ concentrations from 1959 to 2022 in the United States, Canada, Europe, China, and India are 11.2 μg/m³, 8.2 μg/m³, 20.1 μg/m³, 51.3 μg/m³ and 88.6 μg/m³, respectively. Based on the features of the SDE and mean center, the spatial distribution of $PM_{2.5}$ has more spatial variability in the United States, Canada, and Europe and less variability in China and India. The changes in the mean center of the $PM_{2.5}$ concentration exhibit various patterns in each region.

7 Data Availability

Daily $PM_{2.5}$ concentration data at 4011 sites in the Northern Hemisphere from 1959 to 2022 are available at https://cstr.cn/18406.11.Atmos.tpdc.301127 (Hao et al., 2024).

Competing Interests

The contact author has declared that none of the authors has any competing interests.

Acknowledgments

https://doi.org/10.5194/essd-2024-96
Preprint. Discussion started: 2 April 2024

This work was supported by the National Key Research & Development Program of China (2022YFF0801302) and the National Natural Science Foundation of China (41930970). The hourly visibility data are available at from https://mesonet.agron.iastate.edu/ASOS/. The hourly $PM_{2.5}$ data for the United States are available at https://www.epa.gov/aqs. The hourly $PM_{2.5}$ data for Canada are available at https://www.canada.ca. The hourly $PM_{2.5}$ data for Europe available at https://european-union.europa.eu. The hourly $PM_{2.5}$ data for China are available at https://www.cnemc.cn. The hourly $PM_{2.5}$ data for India are available at https://app.cpcbccr.com. The hourly visibility and meteorological data are available at https://www.weather.gov/asos. The monthly global $PM_{2.5}$ dataset for the Atmospheric Composition Analysis Group version V5.GL.04 (ACAG) are available at https://sites.wustl.edu/acag/datasets/surface-pm2-5/). The monthly $PM_{2.5}$ dataset of China High Air Pollutants (CHAP) are available at https://zenodo.org/records/6398971. The monthly $PM_{2.5}$ dataset of Modern-Era Retrospective Analysis for Research and Applications, version 2 (MERRA-2) are available at https://gmao.gsfc.nasa.gov. The monthly $PM_{2.5}$ of the Copernicus Atmosphere Monitoring Service (CAMS) reanalysis are available at https://ads.atmosphere.copernicus.eu/cdsapp#!/dataset/cams-global-reanalysis-eac4.

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